United States Court of Appeals

for the

District of Columbia Circuit

TRANSCRIPT OF RECORD

BRIEF FOR APPELLANT AND JOINT APPENDIX

IN THE

United States Court of Appeals

DISTRICT OF COLUMBIA.

591

No. 8760

ETHYL GASOLINE CORPORATION, *Appellant*,

v.

CONWAY P. COE, AND/OR HIS SUCCESSOR IN OFFICE, COMMISSIONER OF PATENTS,
Appellee.

Appeal from the District Court of the United States for the District of Columbia.

UNITED STATES
COURT OF APPEALS FOR THE
DISTRICT OF COLUMBIA
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PRELIMINARY STATEMENT.

This is the second appeal in this case. The Court rendered its opinion on the first appeal on December 6, 1943. .... U. S. C. A., D. C. ....; 139 Fed. (2nd) 372. That appeal was from a judgment of the Trial Court entered October 27, 1942 dismissing the complaint. Findings of Fact and Conclusions of Law upon which that judgment was based are found at Pages 7-11 of the Appendix in case No. 8408, which by order of the Court becomes a part of the Record on this appeal.

The mandate of this Court was entered on the docket of the District Court on December 21, 1943. Thereafter the District Court filed a memorandum opinion, February 14, 1944 (App. p. 179), in which the Court concluded that the cited patents were "all antecedent to the plaintiff's application" and referring *only* to the cited patents was of opinion that there was no invention, and so found. Based upon that opinion, Findings of Fact and Conclusions of Law were presented to the Court and on March 29, 1944 those Findings, submitted by counsel for the appellee, were adopted by the Court and on the same date he entered judgment dismissing the complaint (App. p. 183). The Record before the Court is now the same as that presented on the first appeal, except the Memorandum Opinion, Supplemental Findings of Fact and Conclusions of Law and Judgment of the District Court, which were entered subsequent to the decision of this Court of December 6, 1943.

STATEMENT OF CASE.

In this Court's opinion on the first appeal it found that the application for patent in this case relates to methods for improving the operation of internal combustion engines, such as automobile engines, involving the use of anti-knock agents; and to products comprising motor fuels and anti-knock materials. The applicant's claims emphasize, particularly, the use in combination of lead tri-methyl ethyl, lead di-methyl di-ethyl, and lead tri-ethyl methyl. The use of lead alkyls in gasoline to minimize knocking was old in the prior patented art but, prior to the present application, reliance had been placed upon lead tetra-ethyl, i.e. lead tetra-ethyl was used in approximately 85 per cent of the motor gasoline sold in this Country and in practically all of aviation gasoline.

In the prosecution of appellant's application for a patent in the Patent Office the Examiner cited and considered the following prior patents (Orig. App. p. 155 to 176 inclusive):

Sullivan et al.	1,938,547,
Voorhees	1,974,167,
Shappirio	2,012,356, and
Midgley	1,573,846.

The Patent Office Examiner originally held that some of these Patents contained subject matter which precluded Bartholomew's claims from being patentable thereover. Upon argument and explanation by counsel the Examiner concluded that the subject matter claimed by the applicant was patentable over these four patents. These four patents were cited thereafter as showing merely the state of the art. He finally rejected the applicant's claims *solely* because the applicant did not present a claim suggested by the Examiner for purposes of interference. Having rejected the applicant's claims upon that ground alone he thereby found that the claims were otherwise, patentable. Upon appeal to the Board of Appeal of the Patent Office that Board sustained the action of the Examiner, and upon rehearing, specifically held that it did not find any ground for rejection other than that stated by the Examiner (App. p. 132).

Thus we find that the Patent Office determined that the applicant's claims were patentable over the prior patents and refused to give applicant a patent *solely* because of his failure to present the claim suggested by the Patent Office Examiner as stated in the opinion of this Court on the first appeal. This negatives any inference that the Patent Office rejected the claims as unpatentable over the prior patents.

Thereafter, suit was filed in the District Court of the United States for the District of Columbia under Section 4915 of the Revised Statute requesting that the Commissioner of Patents be authorized to issue a patent to the plaintiff (App. p. 2). In his answer the defendant contended that plaintiff was not entitled to a patent and relied upon the same grounds given by the Patent Office in rejecting the claims (App. p. 6). The District Court, after trial, entered Findings of Fact and Conclusions of Law, sustaining the action of the Patent Office, i.e. denied a patent to applicant solely because he failed to present the claim suggested by the Examiner (App. p. 7).

Appeal was thereupon taken. The judgment of the District Court was reversed and the case remanded. This Court, in reversing the judgment, ruled that both the Patent Office and the District Court had erred in finding that applicant's claims were unpatentable because of his refusal to present the suggested claim, as the suggested claim did not constitute prior art. This Court thereby disposed of the only reason presented and relied upon by the Patent Office for denying a patent to the applicant.

Upon further consideration the District Court held that the subject matter of plaintiff's claims was unpatentable over the prior patents (App. p. 179). In its opinion the District Court did not show wherein the claims of the Plaintiff lack invention over the four cited patents; nor do the Findings of Fact and Conclusions of Law aid this Court with respect thereto.

In its findings and determination *on the merits, as directed by this Court,* the District Court considered only the testimony and evidence which was before it at the time the first judgment was entered. The latest judgment, from which we here appeal, overruled the experts of the Patent Office who had found that the plaintiff's claims were patentable over the same patents. This, despite the fact that no evidence was introduced to support the judgment of the District Court that the subject matter was unpatentable over those patents. There is, therefore, no evidence in the record to support the Findings of Fact of the Court below as to lack of patentability.

STATUTES INVOLVED.

The pertinent parts of the statute giving jurisdiction are as follows (Rev. Stat. Sec. 4915, 35 U. S. C. Sec. 63):

> "Whenever a patent on application is refused by the Board of Appeals * * *, the applicant, unless appeal has been taken to the United States Court of Customs and Patent Appeals, and such appeal is pending or has been decided, in which case no action may be brought under this section, may have remedy by bill in equity, if filed within six months after such refusal or decision; and the Court having cognizance thereof, on notice to adverse parties and other due proceedings had, may adjudge that such applicant is entitled, according to law, to receive a patent for his invention, as specified in his claim or for any part thereof, as the facts in the case may appear. And such adjudication, if it be in favor of the right of the applicant, shall authorize the commissioner to issue such patent on the applicant filing in the Patent Office a copy of the adjudication and otherwise complying with the requirements of law."

The pertinent parts of the statute having to do with patentability are as follows (Rev. Stat. Sec. 4886, 35 U. S. C. Sec. 31):

> "Any person who has invented or discovered any new and useful art, machine, manufacture, or composition of matter, or any new and useful improvements thereof, * * * not known or used by others in this country, before his invention or discovery thereof, and not patented or described in any printed publication in this or any foreign country, before his invention or discovery thereof or more than two years prior to his application, unless the same is proved to have been

> abandoned, may, upon payment of the fees required by law, and other due proceeding had, obtain a patent therefor."

BARTHOLOMEW'S INVENTION.

We think that it will be helpful at this point to explain Bartholomew's invention, how it arose, and also its novelty and utility over the prior art as disclosed and defined in the Appendix, including his testimony and Exhibits.

The invention relates to methods for improving the operation of internal combustion engines, such as automobile engines, involving the use of anti-knock agents; and to products comprising motor fuels and anti-knock materials. All the claims emphasize, particularly, the use in combination of lead tri-methyl ethyl, lead di-methyl di-ethyl, and lead tri-ethyl methyl. Prior to applicant's invention reliance had been placed, mainly, upon tetra-ethyl as an anti-knock agent.

When appellant's assignor *Bartholomew* first joined the appellant company in 1926 anti-knock was used only to a small degree. At the time he made his invention, lead tetra-ethyl was in extensive commercial use in motor fuel sold in this country and in practically all aviation gasoline (App. p. 15). The premium product is marketed as Ethyl gasoline but, in addition, almost all regular grades of gasoline contain lead tetra-ethyl.

Midgley had invented lead tetra ethyl anti-knock motor fuel. His patent No. 1,573,848, cited in the Commissioner's answer, stated some of the evils of engine knocking, as follows (App. 155, p. 1, lines 44-58):

> "Kerosene, gasoline and the heavier hydrocarbons have the characteristic that, when a combustible gaseous mixture containing one of these fuels and air is burned in an internal-combustion engine while subjected to a relatively high pressure, a fuel knock is produced, *the engine heats rapidly, the efficiency of the engine is reduced and, if the initial pressure is very high, engine parts may be injured.* The highest pres-

> sure at which a mixture may be burned in a cylinder without producing a fuel knock varies with the different fuels and, to some extent, with the temperature, position of spark plugs and other conditions within the engine."

What causes knocking when no anti-knock agent is used, is not yet known (App. 15).

In 1935, plaintiff received complaints from a number of its dealers that engines using fuel containing lead tetraethyl were knocking. Bartholomew and his associates made surveys to determine whether some of the Ethyl gasolines on the market were in fact inferior, but according to the standard laboratory test method then in use, they were not inferior. Finally cars and samples of the gasoline were obtained from the complainants. Bartholomew and his associates found that the fuels which were the offenders were in every respect up to the standard which they had set for the fuel by the then known standard test apparatus. Nevertheless by road tests, they confirmed the dealer's contention that the fuels complained of did produce knocking (App. p. 17).

Bartholomew frequently discussed the problem with *Midgley,* the inventor of Ethyl gasoline referred to above; *Hawley,* assistant director of plaintiff's engineering laboratory; and *Calingaert,* who is director of plaintiff's chemical research laboratory. They were quite concerned about the situation. They made a number of tests with the then standard test apparatus. Some of Bartholomew's associates suggested that the laboratory was using incorrect speed, or incorrect spark advance, or improper temperature, because those are factors that influence engine knocking. It was suggested that a very extensive study of variations of those factors of engine operation be made to see whether it might be possible to find some combination of them which would cause the then standard test apparatus to rate the fuels in accordance with their performance on the road. A very large program of such routine tests was carried out without any result that threw any light on the problem that confronted them; they got nowhere (App. p. 18, bottom, p. 19).

The problem was both serious and complex. Besides the complexities involved, two factors had to be kept in mind throughout. One is cost to the consumer. If too much lead is used, it costs the consumer too much. The other is imposed by an agreement with the Surgeon General of the United States, made many years ago, to limit the amount of lead used in gasoline for civilian purposes to a maximum of three cubic centimeters of lead tetra-ethyl per gallon (3,875 c.c.) of gasoline (App. p. 16, bottom, p. 17).

Bartholomew, knowing that engine design had been materially changed, conceived that the changed design caused imperfect distribution of the fuel to the different cylinders of the engine. It was his conception that the air, being quite light, would be distributed uniformly to the different cylinders, and that, for the same reason, the portion of the gasoline that was vaporized in the manifold would be distributed uniformly to the different cylinders, but that the liquid, i.e. unvaporized, portion of the gasoline would be subject to many influences which would prevent its equal distribution to the different cylinders (App. p. 20, middle).

Bartholomew conceived that one cylinder was getting its normal share of gasoline and lead tetra-ethyl, another was getting less than its share and still another was getting more than its share (App. p. 20, bottom). The prior concept had been that the knocking occurred in the cylinder getting more than its share of gasoline because the unvaporized part of the fuel which went to this cylinder had poor anti-knock properties. Bartholomew conceived the original ideas that the low speed knocking occurred in the cylinder receiving a *lean* mixture of good anti-knock fuel constituents (App. 22), and that the *mixture ratio of fuel to air* had a greater effect on engine knocking than the anti-knock value of the fuel (App. 96). The then known test methods and apparatus were not capable of confirming these concepts. Bartholomew devised new apparatus and methods. With this apparatus he could simultaneously determine the leanness or richness of a mixture in any one cylinder of a multi-cylinder engine and the presence or absence of knocking in that cylinder. With this apparatus and these methods he proved his concepts to be true (App. p. 29).

However, knocking during poor fuel distribution occurred only during low speed operation. Under conditions of good fuel distribution knocking may occur among all the cylinders due to engine compression, temperature of the engine, position of spark plugs, etc. This is called high speed, or uniform fuel distribution, knocking. The periods of engine operation when there is good fuel distribution are much longer than the periods of engine operation when there is poor fuel distribution. During periods of good fuel distribution the engine tends to be quite critical of small differences in anti-knock value of the fuel fed to it, and any deficiency of anti-knock value under this condition of operation is felt quite perceptibly by the engine (App. p. 43).

Bartholomew then conceived the idea that *mixtures consisting primarily of the three lead alkyls, lead tri-methyl ethyl, lead di-methyl di-ethyl and lead tri-ethyl methyl could be made which would reduce the knocking during low speed operation to a minimum while maintaining good operating conditions at high speed* (App. pp. 50-52, 56). His new apparatus and methods proved his concepts. He was thereupon able to construct new curves (App. pp. 49-50), such as E and F, Fig. 4 (App. p. 110), which serve as guides in choosing mixtures to meet specific conditions of engine operation.

The claims at bar define, and are limited to these mixtures as was recognized by this Court in the first paragraph of the opinion on the previous appeal.

Other anti-knock compounds may be used in small quantities without materially varying the effectiveness of mixtures of the three lead alkyls specifically claimed. Lead tetra-methyl is such a "bad actor" that the specification warns the public concerning its use. From a theoretical standpoint it is the lowest boiling of the lead alkyls and so should be the most useful in very cold weather. Bartholomew warns not to use over fifteen per cent of lead tetramethyl in his added material even in the coldest weather (App. pp. 101-102).

The prior patents before the Court are as follows:

(1) Midgley, No. 1,573,846, applied for in April 1922,

(2) Sullivan et al., No. 1,938,547, applied for in March 1932,

(3) Voorhees, No. 1,974,167, applied for in September 1932, and

(4) Shappirio, No. 2,012,356, applied for in November 1932.

The prior patent disclosures are but imperfectly and partially set forth in present Findings Nos. 7 to 10 inclusive (App. pp. 181, 182). These actual disclosures are fully explained by Bartholomew, without contradiction, in his testimony (App. pp. 74-79). None of the prior patents taught or even suggested the combination recited in any of Bartholomew's claims (App. p. 78). None of them taught or even suggested limiting the mixture to the five lead alkyls, i.e. tetra methyl and tetra-ethyl and the three intermediate ones; much less did they teach or even suggest that the three intermediate ones should constitute the primary ingredient. The Sullivan, Voorhees and Shappirio patents do not contain any guide of any kind for choosing a mixture of lead alkyls. They are concerned entirely with the manufacture of organo-metallic compounds, in some cases lead compounds only, without direction as to their selection and use. The Sullivan, Voorhees and Shappirio patents have to do with special ways of manufacturing a wide range of organo-metallic compounds by special treatment of the *gasoline itself*, as distinguished from the usual way of making them separately and then adding them to the gasoline (App. p. 78). In none of them is there any selection proposed of a number for any purpose out of the wide range of the named substances. Midgley does not disclose in his patent the use of mixtures of two or more of lead alkyls (App. p. 79).

The testimony and evidence establish that Batholomew's claimed combination of elements is new; was arrived at by departing from all thought processes, test apparatus and test methods previously known; was unobvious to those skilled in the art; and produces new results of benefit to the public.

STATEMENT OF POINTS.

1. Applicant's claims define invention over the prior patents.

SUMMARY OF ARGUMENT.

This action was brought under section 4915 of the Revised Statutes, 35 U. S. C., section 63, to authorize the Commissioner of Patents to issue a patent to the appellant. The invention, which is the subject of this case, consists of a mixture of certain lead alkyls designed to improve the method of operating internal combustion engines by stopping the knocking in such engines under over-all operating conditions, i.e., throughout the entire range of operating conditions. Bartholomew, the applicant in the Patent Office, the assignor of appellant, when advised that such engines in about the year 1935 when using fuel containing the then used anti-knock ingredient (lead tetra-ethyl) were knocking, set about to correct that condition, which was a serious one to the automobile industry. Many months of fruitless study, experimentation and tests by the then known standard apparatus and methods did not solve the problem. He then conceived the idea that such knocking was due to a character of distribution of fuel and anti-knocks to the engine cylinders which was theretofore unknown. This was an original concept and idea. It was his own. He then devised *new test apparatus and methods* and began his experimentation with the so-called lead alkyls and proved that his concept was correct. He then invented the mixtures of the lead alkyls described and claimed in his application to solve this new problem.

The patentability of the subject matter of the invention over the prior patents was conceded by the Examiner when, asserting that he was acting under Patent Office Rule 96, he formulated and presented the suggested claim. The District Court did not in its first opinion find the applicant's claims to be unpatentable over the prior patents.

In our principal argument we will show to the Court that Bartholomew's invention is patentable over the prior patents and that the Patent Office was right in that respect.

ARGUMENT.

The record shows that the experts in the United States Patent Office concluded that the subject matter of these claims was patentable over the prior patents. The presumption is, therefore, that they are patentable. There is no evidence to the contrary except the disclosures of the four prior patents. The District Court did not hold them unpatentable in its first opinion, and this Court did not hold them unpatentable in its opinion of December 6, 1943. The memorandum opinion of the Court below, entered February 14, 1944 (App. p. 179), upon which judgment was entered on March 29, 1944 (App. p. 183), now holds that the claims are unpatentable over the prior patents. The District Court thus overrules the experts in the Patent Office on the question of the patentability of the subject matter over the prior patents and this, without any decision of the Patent Office to that effect, or supporting evidence other than the prior patents themselves, which lack altogether the disclosure of what is claimed as new in the application.

The Findings of Fact entered by the Court below are substantially the same as those entered prior to the first appeal, excluding those relating to the suggested claim and the addition of Findings of Fact Nos. 15 and 16 relating to unpatentability over the prior patents. Findings of Fact Nos.

11 to 16 inclusive find no support in the record and are clearly erroneous. The fact is that all the evidence shows that the applicant's claims are patentable over the prior patents. The Findings of Fact 11 to 16 inclusive, were prepared by counsel for the Appellee and "mechanically" adopted by the Trial Justice over the objection of appellants, as was stated—and condemned—in *United States* v. *Forness,* 125 Fed. (2nd) 928, 942. (2 C. C. A. January 1942).

It is clear from the record that Bartholomew's combination of elements is new. He arrived at that combination, as a result of his original concepts and by departing from all theretofore known thought processes, test apparatus and methods. The invention as set forth in the claims was unknown to the art. It produces new results, in a new way, and of great benefit to the public. It is, therefore, patentable. See *Loom Company* v. *Higgins,* 105 U. S. 580, 591-592; *Williams Manufacturing Co.* v. *United Shoe Machinery,* 316 U. S. 364, 86 Law Ed. 1537; *Electrons, Inc., et al.* v. *Coe,* 69 App. D. C. 181, 99 Fed. (2nd) 414; *Levin* v. *Coe,* Ct. Appeals D. C. No. 8108, 76 U. S. App. D. C. 347; 132 Fed. (2nd) 589; *Gasoline Products Company, Inc.,* v. *Coe.,* 66 App. D. C. 333, 87 Fed. (2nd) 550, 556.

In this case the appellant finds itself in the novel, but not unpleasant, position of defending the action of the Patent Office in holding that these claims are patentable over the prior art, viz., the four cited patents. Counsel for the appellee finds himself in the most unusual position of attempting to convince this Court that the Patent Office was wrong in holding that these claims are patentable. *He takes this position notwithstanding the fact that the Patent Office has recently issued two patents relating to the same general subject matter, as he frankly told this Court in oral argument on the first appeal, which contain broader claims than those here at bar.* This Court has unquestionably held that this appellant acted within its legal rights by refusing to present the claim suggested by the Examiner for interference purposes, and further held that claim not to be prior art. Had not the Examiner made that suggested claim this appellant would long since have obtained its patent. The appellee having been defeated in his effort to prevent this appellant from obtaining its patent on the theory that the applicant had disclaimed the subject matter of his invention by refusing to make the suggested claim, now shifts his position and rests his case upon the proposition that the claims are unpatentable over the cited patents. The first time he took this position was before the Court below after this Court had rendered its opinion of December 6, 1943.

In the recent opinion of this Court in *Potts et al.* v. *Coe*, —U. S. C. A., D. C.—;140 Fed. (2nd) 470, the Court went to great length in defining what is a patentable invention, and what is routineering or experimentation, in corporate research and test laboratories. In that case the Court held that the appellant's claims represented a combination of ideas disclosed in former patents thereby upholding the Patent Office and the District Court. The facts in that case as regards routineering are entirely different from those here presented. In the instant case the use of all known test apparatus and methods wholly failed to solve the problems with which Bartholomew was confronted. Thereafter, Bartholomew, by his *individual* concepts and achievement, apart from anything known to the art, and after evolving new thoughts as the fundamentals of a problem to be dealt with, made his invention. This was individual inventive achievement. It comes within that class defined by the Temporary National Economic Committee of 1938 as:

> "First, creations which exhibit individual insight * * *." (Footnote, pages 4-5, *Potts et al.* v. *Coe, supra.*)

Any inference that Bartholomew's invention is the result of experimentation in the use of any existing knowledge is completely rebutted in the record. In the circumstances of this case, i.e. having in mind the nature of Bartholomew's invention and the intelligent guides that the application gives in each instance to those skilled in the art, we submit that the claims are entirely definite in form and patentable over the cited patents. *Procter and Gamble Co.* v. *Refining Co.*, 135 Fed. (2nd) 900; *Minerals Separation* v. *Hyde*, 242 U. S. 261, 271, 37 S. Ct. 82, 61 Law Ed. 286; *Motor Improvements Inc., et al.* v. *General Motors Corporation*, 49 Fed. (2nd) 543 (C. C. A. 6th); *Merrell-Soule Co.* v. *Northland Dairy Company*, 28 Fed. (2nd) 924, 927 (C. C. A. 6th).

As this Court stated in its prior opinion, the application related to methods for improving the operation of internal combustion engines involving the use of anti-knock agents; and to products comprising motor fuels and anti-knock materials. Appellee argues, without support from the record, that the claims relating to the method for operating internal combustion engines (process claims 1-4 inclusive) define the ordinary operation of an engine and are therefore unpatentable. The application is replete with statements concerning the improving of engine operation. The most important part of engine operation is the control of the combustion of a mixture of gasoline and air under high compression. Combustion is a chemical reaction. Bartholomew's invention produces a different control of combustion from that produced by the use of lead tetra-ethyl. The accepted mode for claiming a control of a reaction is by a method claim. The Patent Office Examiner accepted these process claims as properly defining the invention.

CONCLUSION.

The appellant's claims are patentable over the cited patents. The Patent Office so held. Neither the District Court in its first opinion nor this Court on the first appeal held to the contrary. There is no evidence in the record of the trial of this case in support of a finding that the claims are not patentable. All of the evidence is to the effect that the claims are patentable. The invention is the individual achievement of Bartholomew and is not the result of corporate laboratory research or step by step progress of an entire group. It is the product of an individual.

inventive ability. It follows that the judgment of the District Court should be reversed.

Respectfully submitted,

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APPENDIX

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IN THE

United States Court of Appeals

DISTRICT OF COLUMBIA.

No. 8760

ETHYL GASOLINE CORPORATION, *Appellant*,

v.

CONWAY P. COE, AND/OR HIS SUCCESSOR IN
OFFICE, COMMISSIONER OF PATENTS,
Appellee.

Appeal from the District Court of the United States for the District of Columbia.

JOINT APPENDIX.

I.

PLEADINGS, DOCKET ENTRIES AND OTHER PAPERS DESIGNATED.

1 Endorsed: Filed Jan. 30, 1941. Charles E. Stewart, Clerk.

In the District Court of the United States
For the District of Columbia

Civil Action

No. 9986

Ethyl Gasoline Corporation, Chrysler Building, 42nd & Lexington Ave., New York, New York, *Plaintiff,*

v.

Conway P. Coe and/or his Successor in Office, Commissioner of Patents, Washington, D. C., *Defendant.*

Complaint for Issuance of United States Letters Patent

Plaintiff, Ethyl Gasoline Corporation, brings this, its bill of complaint against defendant, Conway P. Coe, and/or his successor in office, Commissioner of Patents, and alleges:

I. That plaintiff, Ethyl Gasoline Corporation, is a corporation organized and existing under the laws of the State of Delaware, and has a regular and established place of business at Chrysler Building, New York City, in the County of New York, and State of New York; and that defendant Conway P. Coe, is Commissioner of Patents of the United States and that his office is at Washington, D. C. within the jurisdiction of this Court.

II. The jurisdiction of this Court depends upon the Patent Laws of the United States this complaint being filed under Section 4915, Revised Statutes (Comp. Stats. 9460; U. S. C. Title 35, Section 63), as amended by Act of March 2, 1927.

2 III. The plaintiff, Ethyl Gasoline Corporation, is, by an assignment in writing executed September 1, 1936, and recorded in the United States Patent Office on September 4, 1936, in Liber Z-167, at Page 118 of the Trans-

fers of Patents, the sole owner of the entire right, title, and interest in an application for United States Letters Patent of Earl Bartholomew, for Improvements in Method and Means for Improving Engine Operation, filed in the United States Patent Office on September 4, 1936, Serial No. 99,497, and the improvements disclosed and claimed therein.

IV. The Board of Appeals of the United States Patent Office in a decision rendered on October 15, 1940 and in a clarifying decision dated November 14, 1940, refused to allow fourteen claims of said Earl Bartholomew application Serial No. 99,497.

V. The said fourteen claims in said Earl Bartholomew application Serial No. 99,497, which the Board of Appeals of the United States Patent Office refuses to allow, are as follows:

1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

2. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl and lead tri-ethyl-methyl in which antiknock mixture the lead dimethyl diethyl predominates.

3 2. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of

lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead triethyl-methyl; and lead tetraethyl the concentration of the lead being greatest in the lead triethyl methyl and lead tetra-ethyl.

4. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl, di-ethyl, and lead tri-ethyl-methyl; and lead tetramethyl in a concentration not exceeding fifteen percent.

5. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl lead di-methyl di-ethyl and lead tri-ethyl-methyl.

6. A product comprising a motor fuel and a primary antiknock agent consisting of lead tri-methyl-ethyl, lead dimethyl di-ethyl, and lead tri-ethyl-methyl.

7. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, in which mixture the concentration of lead dimethyl diethyl predominates.

8. A product comprising a motor fuel and a primary antiknock agent consisting of lead tri-methyl-ethyl, lead dimethyl di-ethyl, and lead tri-ethyl-methyl, in which the concentration of lead dimethyl diethyl predominates.

9. A product comprising a motor fuel and an anti-knock material having as its primary anti-knock component a mixture of lead tri-methy-ethyl lead di-methyl di-ethyl, and lead tri-ethyl-methyl, and lead tetramethyl in a concentration not exceeding fifteen percent.

4 10. A product comprising a motor fuel and a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, and lead tetramethyl in a concentration not exceeding fifteen percent.

11. A product comprising a motor fuel and an antiknock agent comprising lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl and in which the greatest lead concentration is in the group consisting of lead triethyl methyl and lead tetraethyl.

12. A product comprising a mixture of a motor fuel, a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetramethyl and lead tetraethyl present in relatively small concentrations.

15. A fuel anti-knock composition comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl and lead tetramethyl in a concentration not exceeding fifteen percent.

16. A fuel anti-knock composition comprising a mixture of lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl with the greatest lead concentration in the lead triethyl methyl and lead tetraethyl.

VI. The defendant, Conway P. Coe, and/or his successor in office, Commissioner of Patents of the United States, by the aforesaid decision of the Board of Appeals in the United States Patent Office, has refused and still refuses to grant letters patent of the United States to the plaintiff, Ethyl Gasoline Corporation, as assignee of said Earl Bartholomew on his application Serial No. 99,497, including said fourteen claims recited in Paragraph V hereof.

VII. The said decision of the Board of Appeals was rendered on or about October 15, 1940, and within six months last past, and no appeal has been taken from said
5 decision of the Board of Appeals to the United States Court of Customs and Patent Appeals, and no such appeal is pending or has been decided.

Wherefore, the plaintiff, Ethyl Gasoline Corporation, prays that this Honorable Court determine that plaintiff, Ethyl Gasoline Corporation, is entitled to the grant of letters patent, on said application Serial No. 99,497, embodying said claims 1 to 12, inclusive, 15 and 16, and that this Honorable Court authorize and direct the Commissioner of Patents to issue to plaintiff, Ethyl Gasoline Corporation, letters patent of the United States, on said application Serial No. 99,497, embodying said claims 1 to 12, inclusive, 15 and 16, upon the filing in the United States Patent Office of a certified copy of such adjudication and upon compliance with the requirements of the law for such cases made and provided.

ETHYL GASOLINE CORPORATION,
By EDWIN R. HUTCHINSON,
Attorney for Plaintiff,
Munsey Building,
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JOE W. MORRISON,
30 Rockefeller Plaza,
New York, N. Y.

H. FRANK WIEGAND,
233 Broadway,
New York, N. Y.
Of Counsel.

* * * * * * * * * *

6 *Answer to the Complaint.*

To the Honorable the Justices of the District Court of the United States for the District of Columbia.

I, II, III, IV, V, VI, VII. Defendant admits the allegations of paragraphs I to VII, inclusive, but denies that the plaintiff, Ethyl Gasoline Corporation, is entitled to the grant of letters patent on the application of Earl Bartholomew, Serial No. 99,497, embodying claims 1 to 12, inclusive, 15 and 16 since it is deemed that the said claims are not patentably different from a claim that was suggested to plaintiff for interference purposes, under the penalty that failure to assert the same would be considered a disclaimer of the subject matter thereof, which suggested claim plaintiff failed to assert and thereby disclaimed; and since it is further deemed that the said claims are also unpatentable in view of the following prior patents:

Sullivan et al.,	1,938,547,	Dec. 5, 1933,
Voorhees,	1,974,167,	Sept. 18, 1934,
Shappirio,	2,012,356,	Aug. 27, 1935,
Midgley,	1,573,846,	Feb. 23, 1926,

7 all for the reasons given in the statement of the examiner in answer to the appeal and the decisions of the Board of Appeals, copies of which will be furnished at the trial.

Profert of copies of the foregoing patents is hereby made.

W. W. COCHRAN,
Solicitor, U. S. Patent Office,
Attorney for Defendant.

February 8, 1941.

I hereby certify that a copy of this Answer was mailed to the attorney for plaintiff on February 8, 1941.

W. W. COCHRAN,
Solicitor.

* * * * * * * * * *

143 *Findings of Fact.*

1. This is an action under Section 4915 R. S. (U. S. C., title 35, sec. 63) in which plaintiff, the assignee of the patent application of Earl Bartholomew, Serial No. 99,497, seeks a judgment which would authorize defendant, the Commissioner of Patents, to issue a patent including claims 1 to 12, inclusive, 15 and 16 of said application.

2. The application was filed in the Patent Office on September 4, 1936, and relates to method and means for improving engine operation and involves the use in the fuel of mixtures of some or all of the following lead alkyls, lead tetra-methyl, lead tri-methyl ethyl, lead di-methyl di-ethyl, lead methyl tri-ethyl, and lead tetra-ethyl.

3. Claims 1, 2, 3 and 4 of the application recite a method of operating an internal combustion engine. Claim 1 reads as follows:

144 1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

4. Claims 5 to 12, inclusive, of the application, recite a product comprising a motor fuel containing the anti-knock lead alkyl compounds. Claims 5 and 12 read as follows:

5. A product comprising a motor fuel and an antiknock material having as its primary anti-knock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

12. A product comprising a mixture of a motor fuel, a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetramethyl and lead tetraethyl present in relatively small concentrations.

5. Claims 15 and 16 of the application recite an anti-knock composition comprising the lead alkyl compounds. Claim 15 reads as follows:

15. A fuel anti-knock composition comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl and lead tetra-methyl in a concentration not exceeding fifteen percent.

6. Claim 1 of the application specifies:

"* * * a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl."

Likewise claims 2, 5, 6, 7 and 8. None of these claims mentions either lead tetra-ethyl or lead tetra-methyl. Claims 2, 7 and 8 additionally require that "lead di-methyl di-ethyl" is the predominating one of the three named. Claim 3 specifies:

> "* * * a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead
> 145 tri-ethyl-methyl; and lead tetra-ethyl the concentration of the lead being greatest in the lead tri-ethyl methyl and lead tetra-ethyl."

Likewise claims 11 and 16. The only claims that mention lead tetra-methyl are claims 4, 9, 10, 12 and 15. Claim 4 specifies:

> "* * * a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetra-methyl in a concentration not exceeding fifteen percent."

Likewise claims 9, 10 and 15. Claim 12 specifies:

> "* * * a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; * * *.",

being in that respect like the other claims that specify those three as the primary ingredients, and claim 12 further states:

> "* * * and lead tetra-methyl and lead tetra-ethyl present in relatively small concentrations.

Claim 12 is the only one of the claims that mentions all of the five lead alkyls lead tetra-methyl, lead tri-methyl-ethyl, lead di-methyl di-ethyl, lead tri-ethyl-methyl, and lead tetra-ethyl. Claim 12 specifies that the lead tetra-methyl shall be present only in relatively small concentration.

7. The following prior patents disclose the use of lead alkyls in gasoline to minimize knocking:

Sullivan et al.,	1,938,547,	Dec. 5, 1933,
Voorhees,	1,974,167,	Sept. 18, 1934,
Shappirio,	2,012,356,	Aug. 27, 1935,
Midgley,	1,573,846,	Feb. 23, 1926.

8. Claims 1, 2, 3, and 4 recite a conventional method of operating an internal combustion engine, and recite also the use of a mixture of certain lead alkyls as an anti-knock agent. The presence of such an anti-knock agent in the fuel does not cause the engine to operate by a method which is different from its method of operation when using previously known anti-knock fuels.

146 9. Claims 1, 2, 3 and 4 are unpatentable.

10. The Patent Office Examiner found to be allowable and suggested to the applicant Bartholomew for interference with one or more other parties, a claim reading as follows:

> "Liquid fuel for internal combustion engines of the otto cycle multicylinder type comprising a mixture of hydrocarbons of such varying volatility and forming such varying air-fuel ratios as to cause material variation in knocking tendency of the several cylinders in operation and containing a small quantity of a mixture of volatile lead alkyls consisting of tetra-ethyl lead, tri-ethyl-methyl lead, di-ethyl di-methyl lead, ethyl tri-methyl lead, and tetra-methyl lead, whereby variation in separate cylinder knocking tendency is eliminated and the anti-knock value of the fuel is raised."

In suggesting this claim, the Patent Office Examiner notified the applicant Bartholomew that he was acting under Patent Office Rule 96 which provides that failure or refusal to make a suggested claim within the time specified shall be taken "as a disclaimer of the invention covered by that claim".

11. The applicant Bartholomew intentionally failed to make the suggested claim and did not seek any extension of the time prescribed by the Examiner and his application has not been placed in any interference.

12. The Patent Office Examiner thereafter finally rejected the applicant's claims solely because the applicant had failed to make the suggested claim.

13. By failing to make the claim suggested by the Examiner, Bartholomew disclaimed the fuel defined by the suggested claim.

14. The fuel recited in the claim suggested by the Examiner to Batholomew contains all of the lead alkyls that are recited in the claims at bar.

147 15. All of plaintiff's claims, except claim 12, are broader in their statement of the ingredients of the anti-knock material than the suggested claim. In this respect claim 12 is of the same scope as the suggested claim.

16. The use of the anti-knock material specified in plaintiff's claims does not eliminate entirely variation in separate cylinder knocking tendency but does reduce said tendency.

17. The selection of the particular lead alkyls and the proportions thereof to be used in the fuel depend on climatic conditions, the particular type of engine being used, and other factors. The selection is made as a result of experimentation.

18. There is nothing critical in the proportions of the ingredients, more or less indefinitely recited in the claims at bar.

19. The claims at bar are not patentable over the fuel defined by the claim suggested by the Examiner to Batholomew.

Conclusions of Law.

1. Plaintiff is not entitled to receive a patent containing any of claims 1 to 12, inclusive, 15 and 16 of the Bartholomew application at bar.

2. The complaint should be dismissed as to all of said claims.

MATTHEW F. McGUIRE,
Justice.

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Judgment.

This cause having come on to be heard and having been tried in open court and argued by counsel for the respective parties upon the pleadings and proofs adduced and submitted to the Court, It is Adjudged this 27th day of October, 1942, that the complaint in this case be, and the same hereby is dismissed with costs against the plaintiff.

MATTHEW F. McGUIRE,
Justice.

Approved as to Form:

....
Attorney for Plaintiff.

* * * * * * * * * *

II.

TESTIMONY DESIGNATED BY APPELLANT.

44 *Evidence on Behalf of the Plaintiff*

Thereupon **Earl Bartholomew** was called as a witness for the plaintiff and, being first duly sworn, was examined and testified as follows:

Direct Examination

By Mr. Wiegand:

Q. Please state your name, age, residence, and occupation. A. Name, Earl Bartholomew; age, 41; residence, 2224 Yarmouth Road, Birmingham, Michigan; occupation, Director of Engineering Laboratories, Ethyl Corporation. 44

Q. That is the Ethyl Gasoline Corporation, which is the plaintiff here? A. Yes.

Q. Would you please take a look at a certified copy of your application, Plaintiff's Exhibit 1? I show you the oath that was originally filed to that application. I believe that is page 25. Is that your signature on there (handing a document to the witness)? A. It is.

Q. You executed the application and it was your application? A. Yes.

Q. Please state what technical education you have had and what some of your experience has been. A. I received a B. A. degree, Bachelor of Science in Mechanical Engineering, Master of Science in Mechanical Engineering from the University of Oklahoma. I was employed for two years as instructor in mathematics and mechanical engineering at the University of Oklahoma; as an instructor in mechanical engineering for three years at Harvard University; after which I started to work for the Ethyl Gasoline Corporation and have been there ever since.

Q. What did you do while you were at Harvard? Did you mention that? A. Well, at Harvard I was an instructor in mechanical engineering, and, in addition, did certain
research work, among the projects being investiga-
46 tion of surface catalysis in combustion chambers of
engines, investigation of orifices with respect to the
effect of orifice geometry on the flow of oil.

Q. When you first went with the plaintiff company did it have an engineering laboratory? A. No. I organized the engineering laboratory.

Q. What kind of laboratory did it have when you first went there? A. Chemical only.

Q. About how many engineers did the plaintiff have in its employ as of about the early part of 1941 or thereabouts, or as recently as you can remember? A. Well, approximately fifty.

Q. When did you leave Harvard to go with the plaintiff company? The year will be enough. A. 1926.

Q. Now, during your experience with the company after you organized the engineering department when you went there, did your company and you yourself particularly have anything to do with any engine development work; and if so, will you just briefly tell us about what that was in the early days and up to the time you were making this invention? A. Well, our research is devoted almost entirely to engine and fuel development. We have participated in the development of the high compression engine; also in the

development of the supercharged engine. We have stood in a peculiar relationship to the automobile industry. Hence, much of the development work which we have done has been in cooperation with the different automobile engineering departments. We have developed engines
47 to be used for the rating of the anti-knock value of gasoline.

Q. Do you happen to be a member of any engineering or technical societies; and if so, will you please state what they are? A. I am a member of the Society of Automotive Engineers and the American Society of Mechanical Engineers.

Q. Have you ever written any papers on engines, fuels, and anti-knock agents or the like? A. Yes, a number of such papers.

Q. Will you please briefly describe the operation of an Otto cycle multicylinder internal combustion engine? A. The Otto cycle engine is known as a four-cycle engine, because the operations in connection with the burning of the fuel and development of power are carried out in four operations. Assuming that my hand represents the piston (indicating)—

Q. That is to say, your fist? A. Yes, my fist. The first of the operations is the suction stroke. The piston moves downward into the cylinder, during which time the charge of air and fuel is drawn into the cylinder from the manifold or from the carburetor, if it happens to be a cylinder that has the carburetor attached directly thereto. On the next upward stroke of the piston the charge is compressed. At or near top dead center, the spark occurs, the charge is burned, driving the piston downward, the pressure exerted on the piston being transformed into power and being transmitted to the crankshaft, constituting the power stroke of the engine. On the final upward stroke of the piston the exhaust products are expelled from the cylinder.

48 By the Court:

Q. By the "charge" I take it you mean the mixture of fuel and air that is sucked in? A. Yes, your Honor.

By Mr. Wiegand:

Q. What causes knocking when no anti-knock is employed? A. Well, that is something that has not yet been satisfactorily explained. We know many of the factors associated with knock, but as yet there is no proof or no explanation of the exact cause of knock which is not subject to a lot of weaknesses.

Q. Is knocking an engine problem or is it a fuel problem? A. Well, primarily it is a problem of engine operation, although the fuel problem does enter secondarily.

Q. Would the same fuel produce knocking in all engines? A. No.

Q. Would the same fuel produce knocking at all times and under all conditions in a single engine? A. No.

Q. To what extent has lead tetraethyl been utilized commercially? A. When I made my connection with Ethyl Gasoline Corporation it was used only to a small degree. Now, however, it is used in approximately 85 per cent of the motor gasoline sold in this country and in practically a hundred per cent of the aviation gasoline.

Q. Had it come into rather extensive commercial use before you made the invention that we are here discussing? A. Yes, it was in quite extensive use at that time. 49

By the Court:

Q. By "ethyl" do you mean gasoline that contains an ingredient of lead—in other words, that lead is one of the ingredients? A. Yes. There is marketed a premium product known as ethyl gasoline, but, in addition, almost all regular grades of gasoline contain the anti-knock agent, of which lead is the active ingredient.

Q. Is that anti-knock lead denoted by the name "ethyl"? A. No, it is not known as ethyl gasoline, but simply as gasoline containing lead. The premium product is known as ethyl, which is a higher quality product.

Q. I was curious as a layman to have determined what the meaning is of the terms "tetramethyl, trimethyl, and dimethyl." I assume that "tetra" means four, "tri" means three, and "di" means two. I can go back to my own experience with Greek and Latin to understand that. But I would like to have the terminology of "ethyl" and "methyl" explained, so I can understand. A. Yes, your Honor. The lead alkyls consist of the element of lead plus alcohol radicals. When I speak of lead tetramethyl, that means that in the molecule there is lead plus four radicals from methyl alcohol or wood alcohol, as it is sometimes known.

Mr. Cochran. With the permission of counsel for the plaintiff, I am permitted to ask the witness a question. May I at this time? I think it will help the very point your Honor made.

50 Mr. Wiegand. Mr. Cochran has an explanation, chemically, and if the witness says it is correct, we agree to it.

(Mr. Cochran handed the witness a diagram.)

The Witness. Mr. Cochran has indicated the structure of them (handing a document to the Court).

Mr. Wiegand. Has your Honor finished your inquiry?

The Court. Yes.

By Mr. Wiegand:

Q. What are the factors, if any, that limit the amount of lead used in anti-knocks? A. Well, one factor is the economic one.

Q. That is, cost? A. Cost. It is desirable to use as small a quantity as possible to achieve a given result.

The other limitation is imposed by an agreement with the

Surgeon General made many years ago to limit the amount of lead used in gasoline for civilian purposes to a maximum of three c.c. of lead tetraethyl per gallon.

Q. What is a c.c., just for the record? A. c.c. is a cubic centimeter.

Q. Do you recall about how many cubic centimeters there are in a gallon, roughly? A. Approximately 3,785.

Q. Now, when comparatively high compression engines came to the fore did you receive any complaints of knocking by engines using fuel containing lead tetraethyl? A. Yes, we did.

Q. About when was that? What year? A. About 1935.

Q. What did you do when those complaints came
51 in from the field? A. We had complaints from a
number of car dealers to the effect that certain ethyl gasolines sold in their territory were satisfactory for operation of their cars and that others were not. We made surveys to determine whether some of the ethyl gasolines were in fact inferior, but according to the standard test method then in use, there was no significant difference. They were all equal, at least to our standard. However, the dealers were insistent that there were differences.

Finally cars were sent to our laboratory. We investigated not only the cars but the fuels about which the complaints had been registered. We found again that the fuels which were the offenders were in every respect up to the standard which we had set for the fuel. Nevertheless we confirmed by road tests the dealers' contention that those fuels in the automobile were substantially inferior to other ethyl gasolines, rating no higher on the accepted testing equipment than the offenders.

Q. Could you interrupt just at that point before we go on and perhaps you could explain, with the aid of this drawing and tell what it is, and whether that shows the standard test apparatus (handing a document to the witness)?

I understand that what you have in your hand is a partial drawing of the standard test apparatus that you had when these complaints came in; is that correct? A. That is right. This is a drawing of the fuel induction system of the standard single cylinder engine of the American Society for Testing Materials used to rate the knocking tendency of the gasolines.

On the drawing we see a note, "Cylinder head of C.F.R. engine." "C.F.R. means Cooperative Fuel Research
52 Committee engine. The Cooperative Fuel Research Committee, composed of representatives of the oil and automobile industries, developed this particular apparatus.

Attached to the cylinder is a manifold containing a 1000-watt heater, and attached to the manifold at the opposite end is the carburetor barrel of the carburetor used to supply fuel to the engine.

In this particular set-up the fuel from the carburetor enters the manifold and flows by gravity downward, if it is not vaporized, and enters the cylinder. So that all fuel, whether it be vaporized or remain as liquid fuel, enters the cylinder and is burned along with the air for the particular charge.

Q. It was such standard apparatus with which you had been making many routine tests from time to time? That is, this was the routine way of doing it, was it not? A. This was the standard and approved method.

Mr. Wiegand. May I offer the drawing in evidence as Plaintiff's Exhibit 2? Is there any objection?

Mr. Cochran. No objection.

(Drawing referred to was received in evidence and marked Plaintiff's Exhibit 2.)

By Mr. Wiegand:

Q. Now, when these complaints came in what did you do? Was it a serious problem or was it just an ordinary run-of-mine problem? A. It was quite a serious problem.

Q. Did you discuss the problem with any of your associates, and would you mind naming a few of them, if

53 you remember who they were? A. Yes, I did discuss that problem with my associates, among whom were Mr. Thomas Midgley, Jr., Mr. Hawley, Assistant Director of the Engineering Laboratories, and Dr. Calingaert, who is Director of our Chemical Research Laboratory.

Q. Do you happen to know whether the Mr. Midgley you have named was the inventor of ethyl gasoline? A. He was.

Q. That is, he got the patent on that some time ago? A. Yes.

Q. Were these discussions that you had just one or two or did you frequently discuss the problem in this committee? A. Yes, we did discuss it quite frequently. We were quite concerned about the situation which was developing.

Q. As a result of those discussions, and I think you said you made a number of tests with your then standard test apparatus, what was the result of that? Did you get anywhere or did you not? A. Well, the fuels, as I stated, were tested first under the conditions standardized by the American Society for Testing Materials, but that showed no differentiation between fuels—which fact showed a big difference in the automobiles. Some of my associates thought that we must be using the incorrect speed or incorrect spark advance or improper temperature, because, as we all know, those are factors which do influence the knocking of engines.

Therefore, it was suggested that we make a very extensive study of variations in these factors of engine operation
to see whether it might be possible to find some com-
54 bination of those factors which would cause the single
cylinder engine to rate the fuels in accordance with
their performance on the road.

A very large program along that line was carried out without any result which threw any light on the problem which confronted us.

Q. Well, stated briefly, it got you nowhere? A. Yes.

Q. Then what happened as far as you were concerned? Did you give up or what happened? A. Well, I was certain that the single cylinder engine would give the same ratings as the car on the road if we had the same conditions existing within the engine which existed in the engines during operation on the road. I was conscious of the fact that in order to develop as much power as possible at high speed, engineers were enlarging the cross sectional areas of manifolds and enlarging the cross sectional areas of carburetors, making the diameters of intake valves larger in the engine—all to permit better breathing at high speeds.

I might mention that along with those changes there had come a change from an updraft carburetor to a downdraft carburetor, simply because the velocities with the enlarged fuel induction system were not sufficient any longer to pull the fuel upward to the manifold. So the updraft carburetor had been replaced by a downdraft carburetor, from which the fuel could flow to the engine cylinders by gravity.

I believed that that change in engine design could have only one effect, namely, imperfect distribution of the fuel to the different cylinders of the engine. It was my conception that the air would be distributed uniformly to the different cylinders, being quite light and homogenous, and that for the same reason that portion of the fuel which was vaporized would be distributed uniformly to the different cylinders. However, that portion of the fuel not vaporized undoubtedly would be subject to many influences which would prevent its equal distribution to the different cylinders.

The proximity of the cylinders to the carburetor, the geometry of the manifold, the length of the manifold, the natural frequency of the manifold as an organ pipe, and many other factors undoubtedly would influence the amount of the unvaporized heavy ends entering the different cylinders.

I believed that the lean cylinder, receiving substantially only the vaporized light ends, was causing the difficulty, because, notwithstanding the fact that the fuel in the lean cylinder—consisting of the lighter fraction of the gasoline, having the highest intrinsic anti-knock value, as contrasted with the rich cylinder, containing a normal charge of fuel, and in addition the heavy ends of lower anti-knock value which under conditions of perfect distribution would have gone into the lean cylinder—contained the fuel having the highest intrinsic anti-knock value, the ratio of air to fuel in that cylinder should be such as to make the knocking tendency quite pronounced, because the tendency for knocking to occur in the cylinder depends to a very large extent on the ratio of air to fuel in the cylinder at the time combustion is initiated. A rich mixture tends to suppress knock, whereas a lean mixture tends to promote it.

So it was my belief that in the cars in which we were having difficulty we were burning in the lean cylinder not
56 the fuel having the composition of the fuel leaving
the carburetor and not the air-fuel ratio of the mixture leaving the carburetor, but a fuel having different composition and a different ratio of air to fuel from that of the mixture leaving the carburetor.

Q. Now, as I understand what you have just said, and summing it up, I get two factors as to the thoughts that you had. I will speak of the first one as it occurred to me in your statement. It was your thought or conception that the different cylinders were getting different kinds of gasoline; is that correct? A. Yes.

Q. Was that a common thought at that time or otherwise? Was there any thought otherwise? A. I do not think there was too much thought about it, although some people that I knew believed that there was a difference in the air-fuel ratio to the different cylinders.

Q. That would be quantity, not kinds; is that correct? A. Quantity, yes.

By the Court:

Q. It would be something other than mere gasoline, which is a combination of the air and the fuel mixed? A. Yes.

By Mr. Wiegand:

Q. The other factor was this. You spoke, if I remember correctly, of some cylinders getting more of the liquid parts of the gasoline than their share? Did you mention that? A. Yes.

57 Q. Just to get it in the record, the volatized part, the vapor portion of the gasoline is known in the trade as the lighter ends of the gasoline? A. Yes.

Q. And the unvaporized portion is called heavy ends? A. Yes.

Q. What was known in the art about the relative values of the inherent anti-knock values of lighter ends as against heavy ends? A. Well, it was generally known and accepted that the lighter ends in the fuel have the superior anti-knock value relative to the heavy ends.

Q. But the cylinder receiving more of the heavy ends was actually not the one that was going to do the knocking, according to your conception? A. That is correct.

By the Court:

Q. May I ask you a question? I understood you to say that when we had knocking as a result of the mixture, it was primarily where you had the so-called heavier ends. A. No; the light ends.

Q. The opinion of the industry and the prior teaching was that it occurred where you had the heavier ends? A. Yes.

Q. And you came to the conclusion that just the opposite was true; that is, that where you had the lighter ends that was true? A. Yes.

Q. But in either case you also had the presence of air? A. Yes.

58 Q. Do I understand you to say that in either case the mixture of air was practically constant? In other words, would you have difficulty or would you be able to determine what you claim to have determined if there was a mixture of air which was constantly varying? In other words, you cannot dismiss the presence of air in the mixture at all? A. That is right. If the distribution of the air were unequal, the problem would be even more

complex than it is. Fortunately, the distribution of air is quite uniform to the different cylinders at all times.

Mr. Wiegand. I think at this point it would be helpful to have Mr. Bartholomew explain some of the things he has been saying in words with the aid of the enlargement of Fig. 2 of the Bartholomew application.

By Mr. Wiegand:

Q. Mr. Bartholomew, you have before you an enlargement of Fig. 2 of your application; is that correct? A. That is right.

Q. Will you please, with the aid of that drawing, explain a little more in detail what you have just been talking about in your last several answers? A. This drawing represents three cylinders of an engine, labeled A, B, and C, a carburetor labeled 11, and an intake manifold labeled 12 through which mixture from the carburetor flows to cylinders A, B, and C.

We have intake valves labeled 14 which open to admit the charge from the intake manifold to the cylinders.

For the purposes of illustration and to indicate the capacity of the cylinders to receive a constant volume
59 of air the pistons labeled 16 are all shown in their
bottom dead center positions instead of at different points in between top and dead center, as they would be in the normal engine.

We have illustrated here what I conceived to be the mechanism of fuel distribution within the engine that was causing the difficulty which I described previously. We represent the mixture of air and vaporized fuel by broken lines, such as surround the Fig. 13, in the manifold.

Unvaporized liquid fuel is illustrated by black dots labeled 17 on the drawing, whose size is indicative of the relative concentration of unvaporized fuel in the mixture.

Now, considering cylinder A one moment, fuel and air leave the carburetor. Some of the fuel is vaporized immediately. We have indicated that by the broken lines in the manifold.

Along with the vaporized fuel, which is mixed intimately with the air, there are drops of unvaporized fuel. That composite mixture starts toward the cylinders.

We have assumed in this particular drawing that cylinder A receives substantially only the vaporized light ends of the fuel. It may be said that it is likely that that would be the case, inasmuch as cylinder A is farthest from the manifold, but that does not necessarily follow. At different speeds it may be some other cylinder which is deficient in receiving the unvaporized liquid portions of the fuel.

We illustrate that cylinder B receives an equal charge of air and of the vaparized fuel, but in addition a certain amount of the unvaporized liquid fuel, which we have indicated by black dots, diminishing in diameter as the drops fall into the cylinder and absorb heat from the
60 surfaces of the cylinder and are consequently vaporized.

The liquid fuel which enters cylinder B we assume to be substantially the liquid fuel in quantity which would enter cylinder B if there were uniform distribution of both vaporized and unvaporized fuel to the three cylinders.

In cylinder C we illustrate the conditions existing in the cylinder receiving not only a normal charge of air and of vaporized fuel but an excess of the unvaporized heavy ends of the fuel, which in the drawing we illustrate as two streams of droplets, as opposed to a single stream of droplets entering cylinder B.

Therefore, it is obvious that we have in the cylinders, when the intake valves have closed, not only a different quantity of fuel but, taken as a whole, fuel of different chemical composition in the three cylinders.

By the Court:

Q. In other words, in cylinder A you have vaporized gasoline plus air, and in cylinder B you have vaporized gasoline plus air and some gasoline in the liquid state, and then in cylinder C you have air plus vaporized gasoline plus a little more of the liquid gasoline? A. That is right, your Honor.

By Mr. Wiegand:

Q. Does cylinder B get about the same kind of gasoline that comes from the carburetor or not? A. Yes. We have illustrated cylinder B as receiving substantially fuel and air in the same proportions as in the charge leaving the carburetor.

Mr. Wiegand. If your Honor please, I do not know
whether you want us to put that large chart in evi-
61 dence or smaller photostats of the exhibit. They
are in Plaintiff's Exhibit 1, but it is rather incon-
venient to handle. I suggest that I put these two sheets
in evidence and that both of them be marked Plaintiff's
Exhibit 3, for convenience.

(Photostatic copies of large chart were received in evidence and marked Plaintiff's Exhibit 3.)

By Mr. Wiegand:

Q. Mr. Bartholomew, up to this point you have been testifying as to your concepts or thoughts in this matter. Did you do anything by way of confirming them, and if so, what did you do? A. Yes, I did set about to determine whether this was a correct conception of what was occurring in the automobile engines on the road.

As I have previously stated, if my conception was correct, then the ratio of fuel to air in the different cylinders would vary widely. Up to that particular time we had no apparatus to permit us to determine whether there were variations in air-fuel ratio among the different cylinders. Therefore, it was necessary to develop apparatus to permit that determination to be made.

I developed a sampling valve, so-called, which we have already seen today.

Q. Maybe you would like to explain that a little more in detail than I did, and I have here a drawing of what I understand to be that.

Mr. Wiegand. I am told your Honor usually adjourns at 12:30.

62 The Court. On Monday we have General Term. I think perhaps it might be wise to adjourn now and reconvene at 1:30.

(Thereupon, at 12:30 o'clock p. m., a recess was taken until 1:30 o'clock p. m., of the same day.)

63 After Recess

The trial was resumed at 1:30 o'clock p. m., upon the expiration of the recess.

Thereupon **Earl Bartholomew**, the witness on the stand at the time of taking the recess, resumed the stand and, upon examination, testified further as follows:

Direct Examination—Resumed

By Mr. Wiegand:

Q. I think that since the Court rose we have switched reporters, so, in order to have the matter straightened out, I think I had better ask the question again.

Mr. Bartholomew, I think you had just begun to explain or discuss, when we recessed for lunch, the tests and investigations you started to make to confirm or find out whether you had the correct thought processes, and concepts, and you were about to explain this sampling device which you now have in your hand.

I should like to ask you, please, to explain what that was and what it was all about; and if you have any drawings or photostats there to aid you in your explanation, will you please refer to them? A. The instrument which I hold in my hand is one designed to be screwed into the cylinder head of an engine in lieu of a spark plug. This instrument is equipped with a small valve in the bottom, operated by a solenoid. In addition, there is a sparkplug incorporated into the instrument, so that it serves both as a sampling valve and as an ignition device.

64 The drawing, here labeled Figure 1, valve for sampling mixture, et cetera, is of a design of this valve which preceded this one. The only difference is in the location of the valve seat itself, which in the design shown on the sketch was about two inches above the bottom of the valve. But in the improved type, which I hold in my hand, the valve is in the bottom of the instrument. The reason why it was moved down there was that I found that when it was up in the body of the valve there was always a little cylinder or pocket of unburned air and fuel which was drawn into the gas sample bottle when the valve was opened, and that always gave a little oxygen in our products of combustion when, as a matter of fact, there should perhaps have been none because of the richness of the mixture.

With this improved type of valve that is in the bottom there is no chance for unburned air to be drawn in in that manner.

This valve was used with associated apparatus installed in the car to obtain samples of the combustion products. In the diagrammatic sketch of the mixture sampling valve and associated equipment, the valve itself is shown on the left-hand side. There is a vacuum pump, which was driven by a motor drawing current from a storage battery within the car, which evacuated a gas-sampling bottle.

You will note a mercury manometer to indicate the degree of vacuum achieved in the gas-sampling bottle. The opening of the valve itself was timed by an auxiliary device which was made and fitted to the distributor. It was arranged to discharge a condenser, shown as 60 microfarads, across the coil in the solenoid coil in the valve at the proper point after the fuel had been burned toward the end of the combustion stroke.

When the valve opened, due to the large difference
65 in pressure, the products of combustion were rapidly
withdrawn from the cylinder into the gas-sampling
bottle. Then the cocks of the gas-sampling bottle were closed, and the gas-sampling bottle was taken to the laboratory for an analysis of the products of combustion.

By methods which are well known, the products of combustion can be analyzed and computations made of the air-fuel ratio. That was done on the different cylinders of the multi-cylinder engines during their operation under conditions of both poor distribution and good distribution.

Q. On the road? A. On the road, yes. It was found that during cold-weather operation and at low speed, there were large variations in the ratio of air to fuel in the different cylinders. That was as much as five ratios. In other words, in the richest cylinder I found that the ratio of air to fuel might be 10 or 11 to 1.

By the Court:

Q. What do you mean by "the richest cylinder"? A. The richest cylinder is the one containing or receiving the largest amount of fuel.

Q. Do you mean C? A. That is cylinder C in our diagram.

By Mr. Wiegand:

Q. On Figure 2? A. Yes, on Figure 2.

By the Court:

Q. That is fuel in any form? A. Yes. After the fuel
enters the cylinder, whether it be in the vaporized
66 state or the liquid state, there is sufficient heat stored
in the piston and the cylinder walls to vaporize the
fuel regardless of the state in which it enters the cylinder.

So, it was found that there were these wide variations in air-fuel ratio, a difference between an air-fuel ratio of 10 or 11 to 1 and 16 to 1, and I found that greatest range was a difference of approximately 50%.

Q. Do you mean that 16 to 1 is 16 parts of air to 1 part of fuel? A. Yes, to one of fuel. So, it was a variation amounting to substantially 50% in the amount of fuel entering the different cylinders. That served to confirm one phase of my original conception.

However, my original conception was more extensive than that. If the original conception was correct, then that difference in the air-fuel ratios among the several cylinders should give rise to differences in knocking tendency; hence, further research was carried out to determine what the variations in separate cylinder knocking tendency amounted to.

At that time there was no method known for determining what the separate cylinder knocking tendencies were; hence, it was necessary to develop means for isolating a given cylinder for the purpose of determining the knocking tendency. Therefore, a device was developed which was used in conjunction with the regular distributor. This device permitted the ignition timing in all cylinders except one to be retarded sufficiently to eliminate all knocking, but the ignition timing in the one cylinder was allowed to remain the conventional timing. Then, after a study had been made in one cylinder, another cylinder could be chosen
to have the ignition timing left normal and the igni-
67 tion timing of the other cylinders retarded. By that
means it was possible, one at a time, to determine
the knocking tendency of each cylinder.

I might add that this device for determining the knocking tendency of each individual cylinder was used simultaneously with the sampling valve and associated apparatus, in order that determinations might be made of the knocking tendency of the cylinder simultaneously with the withdrawal of the products of combustion from that particular cylinder.

After those determinations had been made, I compared the knocking tendency of the different cylinders with the air-fuel ratios, determined as previously described, and found that, in accordance with my original conception, there was a close correlation between the knocking tendency of the individual cylinders and the air-fuel ratios in the individual cylinders. The cylinders which had the greatest tendency to knock were the lean cylinders, which had received the smallest quantity of fuel, and the cylinders having the least tendency to knock were the richest cylinders, which had received the greatest quantity of fuel.

Q. In other words, having in mind the diagram on the blackboard, the cylinder which had the greatest tendency to knock would be A, and the cylinder which had the least tendency to knock would be C? A. That is right; and cylinder B would be in between A and C.

At this point I had confirmed two phases of my original conception, but a third phase remained.

Mr. Wiegand. May I interrupt you just a minute? I should like to offer in evidence the two drawings referred
to by the witness in the last answer, one being of the
68 sampling device, and the other of the hookup for it.
I ask that they be marked as Plaintiff's Exhibits 4-A and 4-B.

The Court. Is there any objection?

Mr. Cochran. No objection.

(Drawing of sampling device was marked as Plaintiff's Exhibit 4-A, and drawing of hookup was marked as Plaintiff's Exhibit 4-B, and they were received in evidence.)

By Mr. Wiegand:

Q. Will you please continue your answer? A. There remained one phase of my original conception which this work had not proved or disproved, and that was the conception with respect to the cause of the variation in air-fuel ratio from cylinder to cylinder.

You will recall that I had conceived of the variation in fuel distribution arising because of the difference in the amount of the unvaporized liquid heavy ends which entered the several cylinders. The investigation which had been

made to date did not prove that the differences in air-fuel ratios arose because of that difference in the amount of liquid heavy ends entering the different cylinders, and the analysis of the products of combustion of conventional gasolines used up to this date did not permit any determination of that particular factor. Hence, I made up a fuel containing only two components, one a quite volatile component, and the other a relatively non-volatile hydrocarbon. These two hydrocarbons varied widely in chemical composition and as such produced products of combustion differing very widely in their composition. Thus an analysis of the products of combustion from the different cylinders permitted not only a determination of the air-fuel ratios
69 in the cylinders but also a calculation of the quantity
of the two components of the fuel which had entered the different cylinders.

By the Court:

Q. Am I correct in understanding or assuming that the experiments heretofore were with relation to the ordinary internal combustion engine and ordinary gasoline, with no leads contained in them? A. That is correct.

Q. Now you are going into the process of examining the products of combustion and then making a determination of the composition of the fuel you used? A. That is correct. The reason for choosing the fuel of the two components which I just described was to give a fuel having light ends and heavy ends of products of combustion widely different composition, containing no lead, in order that a determination might be made not only of the differences in air-fuel ratios among the cylinders, but a determination of how much of the light ends and how much of the heavy ends entered each cylinder.

By Mr. Wiegand:

Q. Just a moment. Let me ask right there, About how many different kinds of hydrocarbons are there in commercial gasoline? Do you know what I mean? A. There are hundreds of hydrocarbons in any ordinary commercial gasoline. They vary widely in their volatility and chemical composition, although the light ends of the ordinary gasoline as opposed to the heavy ends are more nearly similar among members of the group than the two components which I chose for this particular investigation.

Q. Will you please proceed with your answer, now?

70 A. When I had made these determinations, I found that the variations in air-fuel ratio from cylinder to cylinder did, in fact, occur entirely because of the differences in the amount of the heavy component which had entered the different cylinders. Therefore, my original conception with respect to the distribution of fuel in an engine and the consequent effect on knocking in the different cylinders was fully confirmed.

Q. In addition to the apparatus that you have referred to in your last answer, did you devise any other method or apparatus, or both, to enable you to make investigations simulating or imitating the different cylinders of a multicylinder engine operating under actual conditions? A. Yes.

Q. Will you please refer to this drawing and tell me whether that has anything to do with it? If it does, you may use it to aid you in making your explanation. A. I have previously described the fuel induction system for the standard American Society for Testing Materials knock-testing machine. I do not recall the exhibit number which that was given.

Q. The old single cylinder engine? A. The old single cylinder engine.

Q. That was exhibit 2. A. It will be recalled that in that particular fuel induction system, fuel flowed from the carburetor to the engine in a downward direction and was assisted by gravity. Any unvaporized fuel was assisted by gravity in its travel from the carburetor to the engine cylinder. Therefore, all parts of the fuel, whether vaporized or unvaporized, entered the cylinder immediately with the air.

71 We have pointed out that in the multi-cylinder engine there is never equal distribution of the unvaporized portion of the fuel to the several cylinders. It was extremely desirable that, if possible, the standard single-cylinder testing engine be so modified as to permit the simulation of conditions existing in the multi-cylinder engine on the road during poor distribution of the fuel. The fuel induction systems on the sheet which we are looking at at present represent different developments of special manifolds which I made to simulate these particular conditions.

Referring first to the one marked a, it will be noted that inserted between the engine cylinder and the carburetor is a manifold which resembles a gooseneck or a sink trap, and that in the bottom of this trap there is a valve to which is attached a graduated burette.

When this manifold was used, the mixture of air, vaporized fuel, and liquid fuel left the carburetor and traveled downward. However, it was necessary for the direction of flow to be sharply reversed at the bottom of the trap. There was, therefore, a tendency for the liquid fuel not to travel up the right-hand leg of the trap but to flow out through the drain at the bottom.

By the Court:

Q. I take it, then, that the result of this experiment was to get a mixture which was made up primarily of air vaporized fuel; liquid going out at the bottom? A. That is correct.

Q. That was simulating what was going on in cylinder A? A. In cylinder A; that is correct, your Honor.

I found, however, that a further modification was
72 necessary to obtain the exact simulation which was
desired. The velocity through the manifold illustrated in a was still somewhat too high, and there was some tendency to pull droplets of liquid on up this leg into this cylinder. As a consequence, the setup illustrated above c was developed, which is essentially similar to the apparatus

illustrated in a, except that at the bottom of the trap there is an enlargement of the trap. That had the effect of reducing the velocity at the bottom of the trap to a quite low figure, which caused all of the unvaporized liquid fuel to drop out and to be drained from the manifold.

I found that through the use of this equipment I could duplicate exactly the conditions existing in lean cylinder A of the multi-cylinder engine during road operation. But that was not enough. I wished to check the operation of rich cylinder C on the single-cylinder engine. To do that, I recovered the liquid heavy ends during operation under conditions simulating cylinder A and added those in proper proportion to a quantity of the original fuel and then burned that in the engine, either with the conventional manifold which insured all fuel leaving the carburetor entering the cylinder, or in a manifold such as is illustrated in c, but with sufficient heat applied by the heaters to insure complete vaporization.

I found that by that means I was able also to duplicate the behavior of rich cylinder C; and, of course, cylinder B could be duplicated by applying enough heat to the mixture in the manifold to vaporize all the fuel that left the carburetor, so that the quantity of fuel entering the cylinder was
the same as the quantity of fuel per unit volume of
73 air leaving the carburetor, just as in the older type
knock-testing engine.

Q. Let me ask this question: I understand these two diagrams here—that is, the small diagram a and the small diagram c. But what about b and d? A. I beg your pardon, your Honor; I failed to describe b and d.

Q. b and d technically represent the situation with respect to cylinder C? A. b and d were other means for accomplishing the same result. When I found that in the setup in a there was too much velocity to insure the recovery of all the liquid fuel, I developed the setup illustrated in b, which consisted of a cylinder to be attached at the bottom of the gooseneck to provide a greater volume at that point, in the hope that the greater volume there would cause some of the mixture to be breathed from the manifold. That is, I had thought it might be like a multi-cylinder engine—an additional manifold added to the engine. That was partially successful but not nearly as efficient as the enlargement in the gooseneck used in cylinder C.

Q. I understand the reason you gave for the mechanism depicted in this diagram, a, was so that you could duplicate the situation in cylinder A with respect to the fuel mixture, and you found that when you completed this mechanism, due to the velocity with which the fuel mixture was brought in, some of the liquid fuel remained in the bottom of that area. A. Yes.

Q. Then you were uncertain whether it would all remain there or whether some of it was forced out and went on to the other part of the mechanism—that is, through the
74 cylinder head to the engine. So, not knowing absolutely what the situation was, you then developed mechanism as described in diagram c, and you created a larger area down below, making certain that all the liquid fuel would remain in this trap, so to speak. A. Actually, your Honor, b is the next step in the sequence, the cylinder illustrated at the left, which was connected onto the T.

Q. That was the purpose of it? A. That was the purpose of it—to get complete separation.

Q. You did achieve that? A. In c. Actually, before I had had an opportunity to determine that c was completely satisfactory, I designed the setup illustrated in d, the idea being that if the first chamber failed to effect complete separation, the second chamber in the series of three or the third in that series should complete the separation of any liquid fuel that might remain. I found, however, that the apparatus illustrated in c was entirely satisfactory, and although I did try out d, I found that no liquid fuel was recovered from the second and third chambers.

Q. The purpose of the burette was to measure the amount of liquid fuel that you subsequently recovered from the trap; is that correct? A. That is correct, your Honor.

Q. The measurement on there was cubic centimeters? A. Cubic centimeters. I perhaps should have added that simultaneously I measured the quantity of fuel which was fed to the carburetor. Therefore, by this—

Q. (Interposing) You were able to distinguish
75 what was evaporation and what was in the liquid
state? A. That is correct.

By Mr. Wiegand:

Q. What did the investigation using this new apparatus demonstrated here prove to you? Have you mentioned that? A. The proof of my original conception was established by the multi-cylinder engine tests. However, it was a bit inconvenient to have to go out onto the road each time I desired to investigate a fuel. It was much more convenient to be able to go into the laboratory, under the well controlled conditions we have, and regardless of weather conditions outside be able to investigate fuels and antiknocking compounds in the laboratory on the single cylinder engine.

Q. But simulating conditions of multi-cylinder engines; is that correct? A. That is correct.

Mr. Wiegand. At this time I should like to offer in evidence the drawing that has been referred to by the witness in his last several answers. I offer this as Plaintiff's Exhibit 5.

The Court. Is there any objection?

Mr. Cochran. No objection.

(Drawing referred to was marked as Plaintiff's Exhibit 5 and received in evidence.)

By the Court:

Q. What you produced in the nature of a mixture as a result of this experiment, I take it, was a mixture of air plus vaporized gasoline. What did you you discover as the result of the experiment with reference to knocking tend-
ency? A. I found on the single-cylinder engine as
76 well as on the multi-cylinder engine that knocking

was greater in the lean cylinder A than in the lean cylinder C, except that in the case—

By Mr. Wiegand:

Q. (Interposing) Did you mean lean cylinder C? A. Lean cylinder A than in the case of rich cylinder C. However, on the single-cylinder engine I so changed conditions that the conditions with respect to both lean cylinder A and rich cylinder C were set up on the single-cylinder engine, although not simultaneously.

By the Court:

Q. I take it that you found less tendency to knock in the rich cylinder C than in the lean cylinder A? A. There was less tendency for rich cylinder C to knock than for lean cylinder A.

By Mr. Wiegand:

Q. From your discussion so far, one would probably think that the problem you set out to solve was the knocking which occurred during this low-speed, poor distribution period. Is that true? A. No.

Q. Why not? In giving your answer, and with a view to shortening things, perhaps you may refer to page 2, at the bottom, of your application. Do you have a copy of it there? A. I do.

Q. Refer first to that, and then I will refer you to other portions. I think we can then get along a little faster. A. On page 2, the last paragraph, first sentence, I made the statement, quoting:

> "It is commonly known that knocking in automobile engines may occur at either high speed or low speed
> 77 and that a given engine may knock at both high and low speeds."

It follows, therefore, that if we are going to have knocking from the operation, it is necessary to take care of both the low and the high speed conditions of operation.

Q. Is it true that at high speed under ordinary circumstances you have good fuel distribution, so that all the cylinders have the same tendency to knock, speaking generally for the moment? A. Yes. At high engine speed the velocity of the charge through the manifold is greatly increased and is sufficiently high to carry along the unvaporized fuel without the variations in distribution of unvaporized fuel which characterize low speed operation under cold conditions.

Q. This variation in separate cylinder knocking tendency is something that occurs only during a part of the use of the car—a part of the time; is that correct? A. Yes, actually only a very small part of the operating time, in the case of the ordinary automobile.

Q. With the use of the automobile, and where it has good distribution, when there is no difference in the knocking tendencies in the cylinder, that is a greater period in length of time than the other period I just spoke of; is that correct? A. Much greater.

Q. Now, I notice that your application indicates on page 8 that this bad distribution and low speed knocking may occur at speeds as high as 35 miles an hour. I understand that in some states it is illegal to drive at a speed above 50 miles per hour. Now, does not this mean that a person living in one of those states would do most of his
78 driving probably below 35 miles an hour, and he
must be concerned more with low speed knocking than equal distribution knocking? A. I should like to quote from the application on page 8, toward the end of the first paragraph, starting with the sentence that begins, "I find."

> "I find that during conditions existing in the summertime the unequal distribution of fuel described above occurs at substantially constant car speeds up to about 25 miles an hour. Under more adverse conditions, such as winter operation, less heat applied to the intake manifold or the use of less volatile fuel, the unequal distribution of fuel may persist at even higher car speeds."

"At substantially constant car speeds above approximately 35 miles an hour the velocity of the mixture of air and fuel vapor in the intake manifold is usually sufficiently high to entrain the liquid portion of the fuel and distribute it in substantially equal quantities to all the cylinders. Under this condition all the cylinders receive charges which are substantially the same and the charges in all the cylinders have the characteristics of the charge in cylinder B in Figure 2."

Now, whether or not unequal fuel distribution occurs during low speed operation at any particular speed depends, among other things, on the previous operation of the engine. Suppose, for purposes of illustration, that a car is being driven along at 50 miles per hour or at any higher speed in cross-country operation but that it is nec-
79 essary to slow down because of the traffic signals or
vehicles ahead, or for some other reason. The brakes are applied, the car is brought down to a low speed, and is then accelerated again when the road is clear ahead.

During the operation at the high speed, temperature conditions have been built up because of the high speed operation to a point where the manifold, the engine, the radiator, and all the other associated parts of the engine are quite warm. So, when the accelerator pedal is depressed for the acceleration, the heat in the intake manifold absorbed over the previous high speed operation is sufficiently great to vaporize practically all of the fuel, so that under such conditions the period of poor distribution may persist up to only 10 or 15 miles per hour.

On a very hot day or in the summertime on almost any day out in the Southwest or in parts of the South conditions are favorable to good distribution of the fuel, even down to relatively low engine speeds. On the other hand, in the wintertime, when it is quite cold, the conditions of poor distribution may persist up to speeds as high as 35 miles an hour.

Q. Would they persist when the engine warms up under the last conditions you have named? A. The worst condition of poor distribution encountered is soon after starting or immediately after starting in quite cold weather. As the engine warms up, even during cold weather operation, there is a tendency for the poor distribution to be limited to a narrower and lower speed range of operation.

By the Court:

Q. With ordinary gasoline? A. Yes, sir. I refer in all that I have said to ordinary commercial gasoline.

80 By Mr. Wiegand:

Q. I think you have already answered, perhaps, the next question I was going to ask you. If you have, please say so. I think I heard you answer it in part of your last answer. Is there any driving in which the low speed distribution knock plays a very small part? If you have not explained that fully, please proceed to do so. Is there any driving in which the low speed distribution knock plays a very small part? A. I mentioned operation on a hot day. That would be a condition under which poor distribution would play a small part. I also outlined the conditions existing during cross-country driving, where heat is built up in the engine under conditions of high speed driving, and I stated that this heat is available during subsequent low speed operation for vaporizing the fuel.

By the Court:

Q. As I understand your theory, it is this: that you go counter to general opinion previously held or now held by a number of experts in the field, who hold that the cause of knocking is the mixture which has to do with liquid gasoline plus the air. Your theory, as I understand it, is that the knocking you found was caused by a mixture of air plus vaporized gasoline. A. Knocking is due for the most part to the compression that exists in the cylinder at the time the combustion starts. It was my conception that the variation in knocking tendency from cylinder to cylinder resulted from the poor distribution of the fuel.

Q. But the combustible material, taken together with the pressure exerted by the cylinder, is, according to
81 your theory, as I understand it, the air plus vaporized gasoline. A. What I intended to establish was that the difference in fuel distribution—

Q. (Interposing) Taking these cylinders A, B, and C. A. Yes.

Q. In A you have what you call a lean mixture? A. Lean.

Q. That is made up of air and vaporized gasoline. B is made up of air, vaporized gasoline, and some gasoline in liquid form. C is made up of air and gasoline mostly in liquid form. A. Well, the same amount of vaporized fuel as in the others, but in addition—

Q. (Interposing) In addition to that, gasoline in the liquid form? A. All the cylinders may knock. Whether or not all of them knock depends on the compression ratio. But by virtue of the leanness of cylinder A, if there is poor fuel distribution, cylinder A will knock more than cylinder C.

Q. Why? A. Because of the mixture ratio.

Q. What would that be? A. That would be of the order of 15 to 1.

Q. Fifteen parts of air to one part of vaporized gasoline? A. Yes.

Q. Gasoline fuel in vaporized form? A. Yes, your Honor.

I might add at this point that when distribution of fuel is good, we are just as likely to have knocking; but
82 under such conditions the tendency of cylinders A, B, and C to knock will be the same, other conditions remaining constant, because the fuel is distributed uniformly, and the air-fuel ratios in A, B, and C under those conditions become the same.

By Mr. Wiegand:

Q. Is there any condition of operation under which the low speed knock and bad distribution would predominate and, therefore, be the more important to consider? A. Yes. We might consider, for example, the driving done by a physician in a city, in calling on his patients, where he drives a relatively short distance from house to house and never drives at high speed. During the wintertime he would never have the engine warmed up to a point where he would obtain any substantial benefit from the soaking up of heat by the manifold in the higher speed operation which we described previously.

Q. But that is quite a special case, is it, with respect to the use of automobiles generally? A. Yes, as a percentage of the total operation, it is quite small.

Q. Having in mind all those things you have just been saying, which would you consider to be the most important part of the problem you have stated you set out to solve? A. Well, the most important part is to insure good operation during the period of good fuel distribution, because that type of operation constitutes the major part of the operation of the engine.

Q. Have you anything more to say about the problem which you sought to solve, the general background for considering this subject matter—that is, as to good dis-
83 tribution when an engine is warmed up and is very
critical of small changes, or anything of that sort? I do not know whether you get my question. A. I might add—

By the Court:

Q. I take it that you assume that good distribution is typified by cylinder C? Good distribution in the sense that you use the term is typified by cylinder C?

Mr. Wiegand. B, I think, is the good boy, under conditions of good distribution, if you will permit me to testify.

The Court. I only want to straighten it out in my mind.

By Mr. Wiegand:

Q. As I understand it, what cylinder B is getting under conditions of good distribution is depicted in Figure 2? A. That is correct. If we assume that one stream of liquid fuel in cylinder C is transferred over to cylinder A, then each cylinder is receiving one stream of liquid fuel; and they all receive—

By the Court:

Q. (Interposing) Then, of those three cylinders—A, B, and C—B is in the ideal state as far as fuel distribution is concerned? A. Yes.

84 Mr. Wiegand. Will the reporter read my last question which was unanswered?

The Reporter (reading): "Have you anything more to say about the problem which you sought to solve, the general background for considering this subject matter—that is, as to good distribution when an engine is warmed up and is very critical of small changes, or anything of that sort? I do not know whether you get my question."

By Mr. Wiegand:

Q. Will you please try to answer the question? That was left unanswered. A. Yes. At high engine speed or when the engine is hot it does tend to be quite critical of small differences in anti-knock value of the fuel fed to it. Hence, any deficiency of anti-knock value under this condition of operation is felt quite perceptibly by the engine. Therefore, it is important that no step be made in solving any problem which will result in a reduction of the anti-knock value available during the high speed or high temperature operation.

Q. After you completed your experimental and analytical investigations to which you have testified what did you next do to complete your invention, and to aid you in answering that question or to aid you in making yourself clear, I have here an enlargement of your Fig. 4, which is an enlarged

reproduction of some of the curves, colored for your convenience; and also I would like you in answering to refer to your application, and particularly to page 15 near the bottom of your application and page 16 at the top, so that in answering my question, which the reporter will
85 repeat to you, use the chart Fig. 4 that is now on
the blackboard and refer to the specification at the places that I have indicated, if that will aid you.

If your Honor please, that is a duplicate of our Fig. 4 of the application. I think you have the drawings up there as Exhibit 3, and the pages in the specification I referred to are 15 at the bottom and 16 at the top. A. I should like to read from my application, starting at the bottom of page 15, the last sentence.

By the Court:

Q. This is what you did, I take it? A. Yes.

By Mr. Wiegand:

Q. Do the descriptions in your application represent work that you had actually done? You read that application before you executed it? A. Yes.

The Court: I have no doubt about that. My inquiry was directed to what I assumed was the fact that, having in mind these conditions, he did what he is going to read now.

By the Court:

Q. Is that correct? A. That is correct.

(Reading) "I find that the best anti-knock agent is a mixture of lead alkyls which gives the highest and most nearly uniform increase in H. U. C. R. of an engine over the range of operating conditions under which it is used."

I believe, before reading further, I should like to describe this diagram, in order that we may have a better
86 conception of some of the things referred to in the
application.

In the diagram which we have before us there is a vertical scale labeled "Octane number of treated fuel or fraction." That is another way of saying that the vertical scale represents the anti-knock quality of the lead alkyls, which we have shown hereon. As the horizontal scale we have two. One scale is the boiling points at 13 millimeters of mercury pressure in degrees centigrade. The other scale is the number of carbon atoms in the lead alkyl molecule.

By the Court:

Q. In relation to 100? A. No; the actual number.

Q. I see. A. Going back to our consideration of the lead alkyls this morning, it will be recalled that lead tetramethyl contained one atom of lead and four methyl radicals, that is four CH_3 radicals. Therefore, in tetramethyl there are 4 carbon atoms.

Q. I was referring to the perpendicular scale, 84. A. This is with respect to 100.

Q. That is what I thought. A. The octane number scale extends from zero to 100. I do not believe it is necessary to describe it.

Q. No. I wanted to make certain what those figures represented. A. That is correct.

Lead tetraethyl on this chart would be plotted on a vertical line through the 8 carbon atom point.

87 Lead trimethyl ethyl would be through the 5 carbon atom point.

Lead dimethyl diethyl would be through the 6 carbon atoms.

Similarly, the lead triethyl methyl is a 7 carbon atom compound.

After I had done the work which I just described I found it necessary to learn some more about the lead alkyls under different conditions of operation. The only information available with respect to that was certain information; part of the information that was needed for the construction of curve D, which is here labeled a 100 per cent vaporization curve.

By Mr. Wiegand:

Q. The left-hand figure on curve D would be what? A. Point 1 represents the anti-knock value of lead tetramethyl.

Q. That is, the table above the curve gives the compounds or lead alkyls that correspond to the numbers used on the curves; is that correct? A. That is correct. The numbers used on the curves refer to the legend above.

Q. Will you please proceed now? A. Curve D. is a curve representing the anti-knock value of the lead alkyls under conditions of 100 per cent vaporization—that is, when the faulty distribution which we have considered in more detail previously does not exist.

This information with respect to the lead alkyls was generally known, perhaps not as accurately determined as here, but this particular quality of the alkyls was fairly well known.

Our problem is more extensive than that when conditions are such that imperfect vaporization prevails.

88 I had previously determined that the worst condition to be met in service was one when approximately 53 per cent of the fuel is vaporized.

Q. That would represent a condition of poor distribution; is that right? A. Very poor distribution. That is a condition which would exist in about zero weather, immediately after starting of the engine.

Q. Please proceed.

By the Court:

Q. I wonder if I follow this diagram correctly. I do not understand the relationship of the square that you have on the graph, the relationship of the space between 68 and 72 on your perpendicular scale. You have this measurement point here (indicating on diagram). A. Yes.

Q. You have this here (indicating on diagram)? A. Yes.

Q. This curve you are talking about is all in here, probably about 69 or 70 (indicating). A. A little under 70.

Q. What would that represent in relation to this (indicating), if anything? A. I think I had better go back and clarify curve D. We have already explained Point 1. Now let us proceed to Point 2.

Referring to the legend, we find that this illustrates data for trimethyl ethyl. Point 2 has a vertical height here
which when projected over onto the scale indicates
89 that the anti-knock value of trimethyl ethyl under
conditions of perfect distribution would be about,
say, 75.3 or somewhere near that.

By the same token, point 3, representing dimethyl diethyl, when projected over to the vertical scale would have an anti-knock value of perhaps 76.5 under conditions of perfect distribution.

I had found that 53 per cent vaporization represents substantially the worst condition ever encountered with respect to poor distribution, a condition prevailing in very cold weather, near zero degrees Fahrenheit, immediately after an engine is started, or, at least, when there has been no period for the engine to warm up to any appreciable extent.

Curve E was constructed to represent the anti-knock value obtained from the lead alkyls in lean cylinder A under such conditions of poor vaporization. We find that under these conditions lead tetramethyl has a very high effectiveness—it is perhaps about 82.5; that trimethyl ethyl has still quite a high effectiveness, although not as high as tetramethyl—it is about 78.7, perhaps; that as we proceed toward the less volatile alkyls, their effectiveness falls off rapidly.

Dimethyl diethyl has only 72 octane number. Methyl triethyl has only about 69½.

The remainder of the alkyls here illustrated contribute practically no anti-knock values. This is substantially the anti-knock value of the vaporized base fuel itself, for the reason that when only 53 per cent of the fuel has been vaporized, the less volatile alkyls have hardly been vaporized at all, and so only a very small portion of such alkyls enters the cylinder.

We have other conditions of poor distribution, less unfavorable than the 53 per cent applicable to curve E.

We have here a curve marked F, representing con-
90 ditions prevailing when 87 per cent of the fuel is
vaporized. It will be noted here that curve F has
an entirely different shape from curve E.

The different lead alkyls have quite different effectiveness under these conditions from the effectiveness which they have under either conditions that are perfect as illustrated by curve D or the 53 per cent vaporization as illustrated by curve E. Here lead tetramethyl is down somewhat with respect to trimethyl ethyl. Dimethyl diethyl, illustrated by point 3, is about on a par with trimethyl ethyl. As we proceed toward the less volatile alkyls, their effectiveness tends to fall off slowly.

Lastly, I made determinations of the behavior of the lead alkyls under a wide range of conditions, in between 53 and 87 per cent vaporization, and in between 87 per cent vaporization and 100 per cent vaporization. I did not include those in this drawing, because they would tend to confuse the illustration of the principle employed.

By Mr. Wiegand:

Q. Now, did you or did somebody under your direction make actual tests of these individual lead alkyls to get the data for this curve sheet that you have been testifying about? A. Yes, sir.

Q. What test apparatus or test method did you use in that connection? Is it anything that we have here that you talked about? A. Yes. I used the single cylinder engine equipped with the—

Q. That is Exhibit 5? A. The one illustrated
91 about c.

Q. That is right. A. I checked that data also by actual car operation to make certain that there was close correlation between what was determined on the single cylinder engine and what could be expected in road operation.

Q. So you now had information as to the behavior of the various lead alkyls there depicted on that Fig. 4 used individually under conditions of good distribution and under conditions of poorer distribution and then still poorer; is that correct? A. That is correct.

Q. The poor or poorer distribution curves would represent what you have heretofore talked about as cylinder A? A. That is correct. I might add to that that curve D would be indicative of the conditions existing in cylinder B of our previous diagram, which received a normal charge.

Q. Did that information that you got from that test data and which you have depicted or visualized on Fig. 4 aid you in any way in finding the mixture that you wanted to solve your problem? A. Yes, it did. In fact, those curves enabled me to determine the type of compounds or of lead alkyls which I have in a mixture to cover the wide range of operating conditions to be encountered in service.

Q. And with which you were concerned? You were not concerned with merely part of operating conditions, but the entire, over-all operating conditions? A. The entire range, that is correct.

92 Q. That enabled you to go to your further test, which I will inquire about in a moment, but suppose you lacked such direction; what have you to say about the time or the infinite permutations or combinations you would have had to make otherwise? A. Well, without such direction it would have been a problem of endless testing. There is an infinite number of combinations of different percentages of lead alkyls that might be made up. It would have been necessary to test all of those combinations, a program which would have extended over a period of many years.

Q. Before we proceed with just what you depicted there and your further test to confirm that, I think you spoke of some data as to curve D having been known before you did this, but what about curves like F and E, showing values of individual lead alkyls under conditions of poor or poorer distribution? Was that known before you did this, so far as you know? A. To my knowledge, curves of the type of E and F had not been constructed previously.

Q. And, so far as you know, you had not seen any data published by anybody relating to that sort of thing, had you? A. No such data.

Q. I think we can go back to the proposition or the statement you made, if I understood you correctly, that with this family of curves of individual properties of anti-knocks under these conditions of good and poor and poorer vaporization, you were able to project your thought as to the proper mixture to select. Would you please go on and answer that? A. I should like to continue reading my application.

93 Q. Do it in any way you like. A. I believe that states it as well as I can.

Q. I think that I can probably help you. You want to refer to the parts that have to do with your problem; is that it? A. I want to refer to the selection of the alkyls. I now start reading at the bottom of page 15:

"I find that the best anti-knock agent is a mixture of lead alkyls which gives the highest and most nearly uniform increase in H. U. C. R."—

I will pause for a moment to define "H. U. C. R." That is a provision for highest useful compression ratio.

Q. What relation does that have to knocking tendency? A. The higher the highest useful compression ratio, the better the anti-knock value.

Q. Or the less the knocking? A. Or the less the tendency to knock.

Q. All right. A. (continuing) "of an engine over the range of operating conditions under which it is used. I find that this mixture should contain in substantial quantity a lead alkyl which has high inherent anti-knock effectiveness over a wide range of engine speeds and as low a boiling point as possible consistent with this requirement, and that the 7 carbon atom compound, lead triethyl-methyl, best meets these conditions."

That is No. 4 on our diagram.

"I find that this mixture should also contain in substantial quantity a lead alkyl which has a low boiling
94 point, relatively high effectiveness under conditions of poor distribution and not too low inherent anti-knock effectiveness over a wide range of engine speeds, and that the 5 carbon atom compound, lead tri-methyl-ethyl, best meets these conditions."

The 5 carbon atom compound is No. 2 on our chart.

Q. The chart Fig. 4? A. Yes, Fig. 4.

"The 6 carbon atom compound, "which is illustrated by point labeled 3 in Fig. 4, "has a relatively low boiling point and a relatively high and substantially uniform anti-knock effectiveness over a wide range of engine speeds and I find that this compound should be used in the mixture in substantial, or even predominant, proportions."

Q. According to your invention, what is the primary anti-knock, if I make myself clear? A. Yes.

Q. What did you pick as a primary anti-knock? A. I picked as the primary anti-knock agent a mixture of the three alkyls which I have just named: In other words, trimethyl ethyl—

Q. That is No. 2? A. No. 2 of Fig. 4; dimethyl diethyl, which is No. 3; and triethyl methyl, which is No. 4.

Q. Suppose we were wholly concerned with periods of poor distribution. Didn't I understand your testimony to be that No. 1 would have the best anti-knock value under this condition of very poor distribution? A. That is correct. For conditions of extremely poor distribution, lead tetramethyl, illustrated by point 1 in Fig. 4, has the highest effectiveness.

95 Q. Well, now, we will come back to that later, particularly in some of the other questions I have to ask you later on.

As I understand it, in your invention the primary ingredient comprises the mixture of 2, 3, and 4 as identified on Fig. 4? A. That is correct.

Q. After you had this information and curves here you said a moment ago that that enabled you to project the mixtures that would be the best in your thought. Did you test and confirm your thought in that direction? A. Yes, quite extensively.

Q. Those tests confirmed what you thought or deducted from your information that you got from Fig. 4? A. They did.

Q. And you did not test all of the possible infinite varieties of the combinations of those elements enumerated there? I mean mixtures of them. A. No. That was not necessary.

The Court. I take it that those lead alkyls are all anti-knock agents in essence, and what you have is a composition of lead alkyls that eliminate knock under conditions testified to, and what you have determined to be the essence of the invention is the specific combination of these lead alkyls of 45 per cent, 30 per cent, and 25 per cent?

Mr. Wiegand. To answer your Honor's question directly, some of the claims—indeed I think the vast majority of them—define that the second, third, and fourth there
96 are the primary anti-knocks.

The Court. The primary anti-knock is the dimethyl diethyl, in the sense that he uses that in the greatest percentage, 45 per cent; 25 per cent of trimethyl ethyl; and 30 per cent of triethyl methyl.

Mr. Wiegand. But, in any event, the middle three predominate. There may be a little variation in there.

The Court. He gets the ideal result, as I understand it, under the percentage herein stated.

Mr. Wiegand. Did you want the witness to answer that?

By the Court:

Q. You get the best result by using 45 per cent of dimethyl diethyl, 30 per cent of triethyl methyl, and 25 per cent of trimethyl ethyl? A. Yes, considering the whole range the year round.

Q. Under driving conditions as a whole, you get the best results from using that composition? A. On the average, yes.

By Mr. Wiegand:

Q. Will you refer to your specification again? I would like to refer you to certain places there and ask you whether they have any bearing or relation to the problem you were solving. Will you turn to the first sentence of the last paragraph of page 2, and then on page 3, where you state what the object of the invention is, and then the top of page 4, where you state, "Where the engine may be operated with wide variance," and then on page 16, where it begins, "to meet the widest normal range," and also on page 17, the second paragraph, second sentence, and page 18, where it says, "I have found that when," and so forth?

97 As you take those up you might just indicate in each instance where and in what place you are reading, and what relation it has, if any, with respect to the problem you were solving and did solve, according to your testimony. A. Considering first page 2, the last paragraph, the first sentence:

"It is commonly known that knocking in automobile engines may occur at either high speed or low speed and that a given engine may knock at both high and low speeds."

Q. Did you want to explain that, if you have not already?

The Court. I think that is common knowledge.

Mr. Wiegand. I think that has been pretty well cleared up.

By Mr. Wiegand:

Q. Did you want to go on any more there on that page or page 3? A. I think I shall go to page 3.

Q. All right. A. To the third paragraph, which I shall read:

"The objects of the present invention are to so direct this correlation that improved performance may be obtained."

I think I had better read the paragraph preceding that.

Q. Suppose you do that, if it is necessary or desirable to get the meaning. A. "The characteristics of engines, base fuels and admixed anti-knock compounds are interdependent in the determination of the occurrence and
98 severity of knocking. I have found that the different
lead alkyl anti-knock compounds have certain different properties which, as related to certain properties of base fuels and features of engine design, form a broad background for improving engine operation. I have also found that these fundamental characteristics may be correlated to improve engine performance.

"The objects of the present invention are to so direct this correlation that improved performance may be obtained under the varied conditions of operation of low duty and high duty internal combustion engines. By high duty engines I mean engines capable of operating at high volumetric efficiency over a wide range of rotative speeds."

I do not know whether much explanation of that quotation is required or not. It simply means that there is a broad problem to be met.

Q. Then refer to the top of page 4 and see if you find anything there that has any bearing on this problem that you were solving. A. I should like to read the sentence in the first paragraph, starting:

"Where the engine may be operated with wide variance in some or all of these conditions the anti-knock mixture may be chosen in accordance with my invention so as to give a high average engine performance throughout."

Q. Now, on page 16, a little below the middle. I think the sentence starts, "To meet the widest," and so forth. I think that is what the Court referred to a while ago, 99 is it not? I did not have it before me then. You might put it in the record. It is not very long. A. About two-thirds of the way down in the first paragraph:

"To meet the widest normal range of operating conditions my preferred mixture consists, on a volume basis, of 25 per cent lead tri-methyl-ethyl, 45 per cent lead di-methyl di-ethyl and 30 per cent lead tri-ethyl methyl."

By the Court:

Q. Continuing further, what is meant with reference to so-called halides?

By Mr. Wiegand:

Q. Will you please explain that to the Court? A. The organic halides, specifically of bromine and chlorine, are used at the present time with lead tetraethyl in order to have the products of combustion of the lead, as lead bromide and lead chloride—

By the Court?

Q. Those are salts? A. Yes, they are organic salts. In the reaction they combine with the lead to form the lead halides—that is, lead bromide and lead chloride—which are vapors and as such are exhausted from the engine. If the halides were not used, the lead would burn to lead oxide, which is a solid, and would remain in the engine and cause trouble.

By Mr. Wiegand:

Q. I think the next one is on page 17, second paragraph, second sentence. A. Page 17, second paragraph,
100 second sentence:

"The proportions of the methyl-ethyl lead alkyls, and of the tetra-methyl and tetra-ethyl compounds if the latter are used, may be varied to meet different ranges of climatic conditions, operating conditions and engine design."

Previously I stated my preferred mixture which I had found to be best to meet the average conditions existing throughout the year, considering all types of engine operation.

However, if it were known that a given engine operation would deviate from that average, as, for example, toward lower speed driving or driving under cold weather conditions, the mixture could be varied to give a better result under those specific conditions.

Likewise, if it were known in advance that great emphasis were to be given to high speed operation or operation under conditions of good distribution, the mixture could be varied in the directin to give more emphasis and consequently better results under those conditions.

Q. Now, I think the last reference is on page 18, in the paragraph in the middle, beginning with, "I have found." I think, in view of your previous discussion of cylinders A, B, and C, you can first quote that and then, as briefly as you can, explain what that means, as a sort of summary. A. Reading from the application:

"I have found that when using the mixture or mixtures described above, under the conditions of imperfect fuel distribution described with reference to cylinders A, B and C,
that the knocking of the cylinder A is reduced to
101 a minimum consistent with good engine performance
over the entire range of operating conditions."

Q. Now, if you solved the condition of knocking during poor distribution, would not that solution take care of the entire problem that you were seeking to solve? A. No, not at all.

Q. Why not? A. I think I could best explain that by saying—

Q. You are now looking at the chart Fig. 4? A. Yes. Looking at Fig. 4, if the only problem to be solved were one where poor distribution exists, one would be led to the use of lead tetramethyl and nothing else, because, as shown by curve E, point 1, under such conditions lead tetramethyl has the highest effectiveness.

Q. That is, for the poor distribution conditions you would pick lead tetramethyl? A. That is right.

However, under conditions of good distribution, lead tetramethyl would be very poor, as will be noted by reference to point 1 on curve D, which represents the 100 per cent vaporization condition. Lead tetramethyl is worse than any of the others under that condition.

If we should solve the quite poor distribution problem without any reference to other conditions of operation, we should have very poor operation under the other conditions.

Q. I think in your application you say—I won't stop now to find the page and line; perhaps your recollection will serve you—where you use tetramethyl at all you said that you would never go over 15 per cent. Do you
102 recall that being in your application? A. Yes. If
tetramethyl is present at all in my mixtures, it should be present in very small amount.

Q. Notwithstanding that it would be the best curative or remedy, if I may put it that way, if you were merely concerned with non-uniform or poor distribution conditions; is that correct? A. That is correct.

By the Court:

Q. I understand it, then, that no matter what the distribution situation is, with respect to the average car that drives into the filling station, if he uses this composition, the base being 2, 3, and 4—that is, the dimethyl diethyl, the trimethyl ethyl, and the triethyl methyl, in the percentages that you have stated—you would come pretty close to solving the knocking situation? A. That is correct, over the whole range to be encountered.

By Mr. Wiegand:

Q. Having in mind also, if I may put it in a way that a little darkey did once—he wanted to get the mostest for the leastest—if you were permitted to have enough of any one of these, you would get rid of your knocking.

The next question may take a rather long answer. Does your Honor want to proceed?

The Court. I think so.

By Mr. Wiegand:

Q. I think you testified this morning, or a while ago, about your thought about knocking in multi-cylinder
103 engines, and you were talking about this electromagnetic device, and then your single cylinder test and method of using the gooseneck that had the four figures, a, b, c, and d, and then you had drawn certain conclusions and deductions from that before you proceeded with this Fig. 4 investigation.

Up to the time that you reached Fig. 4, and you had made all these analyses and projected your thoughts and confirmed them as to what was causing the difficulty, did you publish results of them and show any of these things to your associates interested in such things? A. Yes. At the annual meeting of the Society of Automotive Engineers in Detroit in January, 1938, I read a paper entitled, "Carburetion, Manifolding, and Fuel Anti-Knock Value," which had been written jointly by two of my associates and myself.

In this paper I stated the principles which I have previously described with respect to distribution of fuel within the engine and the resultant effect on the separate cylinder knocking tendency.

The Court. This is being offered for what purpose?

Mr. Wiegand. If your Honor please, the purpose is this, if you do not mind my testifying in advance of the witness.

The Court: I am merely asking the purpose.

Mr. Wiegand. I want to show that as late as the date when he published these conceptions of his about the cylinders getting different kinds of gasoline, you will see that Mr. Blackwood said, in polite language, "I do not agree with you," if you will turn over to the back of the publication. He still persisted in things people
104 believed away back when he did that work.

The Court. I thought you were going to introduce this to cast some reflection on the interference proceeding.

Mr. Wiegand. No. We are not concerned in proving our date of invention.

The Court. That is what I wanted to make sure of.

Mr. Wiegand. As a matter of fact, I do not know whether that man is in interference or not. I do not know whether he got an application or not.

Mr. Reporter, will you read the last answer of the witness?

(The last answer was read by the reporter, as recorded above.)

By Mr. Wiegand:

Q. Will you continue from that point on? A. However, I made no mention of the lead alkyls in this particular paper.

Q. That is, you just described there your sampling apparatus and the new single cylinder method of testing and your conclusions with respect to fuel and distribution, without mentioning alkyls? A. Yes.

Q. Was that accepted by your associates there or was there anyone who dissented from your views?

The Court. How material is that?

Mr. Wiegand. I wanted to explain just what it means. It is engineers' language.

The Court. He can if he wants to, but it is not helpful to me. I do not think it is. As I understand it, what he
105 is testifying to now is that he had this theory in
mind and that he wrote a paper setting forth his theory, and there was some discussion about it, but he did go further and go into the business of the composition of the matter that eventually led to a resolution of the problem.

Mr. Wiegand. What I wanted to bring out in a further question beyond the one that I have put so far is this: that if he had continued with what the Standard Oil man was saying then, he would not have brought about this solution.

The Court. There would not have been any solution.

Mr. Wiegand. If he had persisted in that thought process, he could never have made the invention we have here.

The Court. That is what I understand. In other words, he was the voice of one crying in the wilderness. They did not agree with him. The Standard Oil man did not agree with him. It was a revolutionary idea.

Mr. Wiegand. That is entirely satisfactory.

The Court. That is the way I have it.

By the Court:

Q. Is that correct? A. That is correct.

The Court. Then he proceeded with his experimentation.

Mr. Wiegand. Do you have any objection to my offering it? It is not offered for the purpose of proving the date of his invention.

Mr. Cochran. I do not see what materiality it has.

The Court. It is merely, as I understand, supplementary to what the witness has already stated: that he had different theories that were entirely radical in the
106 industry with respect to what was causing the difficulty. As I understand it, it was a question of lean mixture and rich mixture. He read his paper and performed some experiments antecedent to that, and then he went ahead on the basis of the theory that he had, and the result is what is alleged now to be invention involving the composition of the mixtures.

Mr. Cochran. I do not object to it.

Mr. Wiegand. May it go in, just for the sake of the record?

The Court. I will be very glad to have it.

(Publication entitled, "Carburetion, Manifolding, and Fuel Anti-Knock Value," was received in evidence and was marked Plaintiff's Exhibit 7.)

The Court. Let us adjourn until tomorrow morning at 10 o'clock.

(Thereupon, at 3:30 o'clock p. m., an adjournment was taken until tomorrow, Tuesday, May 12, 1942, at 10 o'clock a. m.)

110 **Earl Bartholomew**, the witness under examination at the time of adjournment on yesterday, resumed the witness stand and testified further as follows:

Direct Examination—Resumed

By Mr. Wiegand:

Q. Mr. Bartholomew, I notice that the Examiner in his Statement, paper No. 14 in your application file, and specifically at page 3 of it, said:

"The invention disclosed is concerned with improvement of motor fuels such as gasoline with respect to its tendency to knock in multicylinder internal combustion engines by addition of lead alkyls as anti-knock dope. Applicant proposes use of certain mixtures of lead alkyls as anti-knock."

Dropping down a paragraph, he said also:

"Applicant's mixture is one of adjusted volatility and anti-knock effect thru use of mixtures of lead methyl and ethyl derivatives such as lead tetraethyl, lead tetramethyl and intermediate alkyls, lead tri-ethyl-methyl, di-ethyl di-methyl and tri-methyl-ethyl."

Now, is that an adequate description of your invention? A. No. My invention was directed toward improving engine operation throughout the range of conditions encountered in service, and not merely toward improving operation during conditions of imperfect fuel distribution. 111

By the Court:

Q. What do you mean by "improving engine operation throughout"? A. As I stated yesterday, there is for the average engine a much greater period during which distribution is good than there is a period during which distribution is poor. It is essential to have satisfactory operation not only under conditions where poor distribution of the fuel obtains but also under conditions where good distribution characterizes the operation.

Q. Well, is not good distribution of fuel something that is purely mechanical? A. Well, good distribution of the fuel depends on a number of factors of engine operation—the speed, the temperature——

Q. Would it not depend primarily, in essence, first of all, on the engine itself and on the structure of the engine? In other words, if you had an engine that was not in good physical shape, so to speak, the fuel would be acting under a poor medium—that is, it would be operating in a poor operating medium? A. That is true. The efficiency of distribution which will occur under any given set of conditions does depend on the design of the engine. It would be possible to design an engine for low-speed operation
only which could be made to give very good distri-
112 bution at low speed, if there were no desire to have
operation at high speed.

By Mr. Wiegand:

Q. Will you please turn to page 3 of your application? Down at the last paragraph on that page it begins with this sentence:

"I have found that mixtures of the three methyl-ethyl lead derivatives may be used, with or without other lead alkyls, with the various types of gasoline marketed to obtain better results in variable-speed, low duty and high duty engines than are obtainable under a wide range of operating conditions with any single lead alkyl."

Just what did you mean by that? Did it have anything to do with economy? A. In the last analysis, it is a problem of economy. Lead is the active or basic ingredient in all of the lead alkyl anti-knocks. With a mixture of the three lead alkyls just named, I can get for a given use of lead a better result with the mixture than I can obtain with any single lead alkyl containing the same amount of lead.

Therefore, for a dollar it is possible to purchase more anti-knock value in the form of the mixture than in the form of any single one of the lead alkyls. Or, to put it in another way, if a limit of three c.c. per gallon is imposed, it is possible to accomplish with a mixture, properly chosen of the three named lead alkyls a better effect than can be obtained by the use of any one of them or any one lead alkyl.

Q. Now, turning back to the Examiner's Statement again
that we started to look at this morning—let us look
113 at page 3 of the Examiner's Statement, which is page
66, as marked down in the right-hand corner of Exhibit 1—and there the Examiner was saying, at the bottom of the page:

"In ordinary carburetion of an air-fuel mixture comprising a mixture of air, fuel vapor and unvaporized suspension therein of liquid fuel particles, inertia effects upon the suspended particles of variable distances the mixture travels to the several cylinders, variable speeds of operation as rapid acceleration, starting, stopping as well as temperature variation, will give variation in the fuel-air charge actually received by individual cylinders. This variation results in different knocking tendencies of the several cylinders."

Can you find that on page 3? Do you have a copy there? A. I do not have a copy of the Examiner's Statement, but I heard it.

Q. Will you look at this (handing a document to the witness)? A. All right.

Q. Is that statement correct? A. Yes, it is.

Q. Now, the Examiner's Statement proceeds in the next succeeding paragraph:

"By using a mixture of lead alkyls of variable volatility and anti-knock value to be added to the fuel, applicant contends that this condition is improved due at least in part to better distribution of anti-knock under such variable conditions."

114 With your mixture as described and claimed in your application, do you increase the variation in separate cylinder knocking tendency or do you decrease it? A. I increase it.

Q. So you do exactly the opposite of what the Examiner said you do in this instance? A. That is right.

Q. If one wanted merely to decrease the variation in separate cylinder knocking tendency would he use your invention? A. No.

Q. Is there any one of the five alkyls we have been talking about or any mixture of them that will decrease the variation in separate cylinder knocking tendency. A. Under a limited range of poor distribution—that is, a range where distribution is only slightly bad—the use of lead tetramethyl alone would result in a slight decrease in the variation of knocking tendency among the different cylinders. Under the same conditions the use of lead trimethyl ethyl would leave the variation in separate cylinder knocking tendency substantially the same.

Q. That is, it would neither decrease nor increase it? A. That is right. The use of any other of the five named lead alkyls would result in an increase in the variation in separate cylinder knocking tendency.

Q. Now, I may have asked this yesterday, but just for the sake of being sure: Under conditions of perfect distribution, all of the cylinders have the same tendency to knock; is that correct? A. Other conditions remaining the same.

115 Q. Now, I call your attention to the top of page 3 of the Examiner's Statement, where the Examiner started, "The proposed claim under rule 96 is copied:" and then he quotes a claim. Do you have that before you? A. yes.

Q. You have read that now, have you, and you have read it before, of course? A. Yes.

Q. On page 4 of the Examiner's Statement the Examiner said, at about the second full paragraph on that page:

"As the examiner understood the invention, a claim as copied above was formulated to express the same and proposed under rule 96 to applicant and other parties."

Then he states further in regard to that same claim, dropping down to the next paragraph:

"Applicant argues that the proposed claim is inoperative and he has no basis therefor, at least in the functional clause,

"'whereby variation in separate cylinder knocking tendency is eliminated——.'"

Did the claim formulated by the Examiner express your invention. A. No, it did not.

Q. The Examiner refers to that limitation, "whereby variation in separate cylinder knocking tendency is eliminated."

Do you say anything in your application concerning the elimination of variation in separate cylinder knocking tendency?

116 I refer you to page 12, so that we may shorten things here. On page 12 there is something about the ideal solution. Can you get that? A. Yes, in the second paragraph.

Q. What did you say there? Just read it into the record quickly. A. "The ideal solution of this problem would be to distribute the anti-knock agent in accordance with the needs of each cylinder but this is impractical."

Q. If you have not already explained, why is it impractical? A. That would require some type of mechanism to anticipate the anti-knock requirements of each cylinder before the need actually arose and to furnish to each cylinder immediately the requisite amount of anti-knock value to take care of the requirements of each cylinder and no more.

Q. Does your application disclose any such apparatus? A. No.

Q. Now, do you say anything else in your application—and I will again call your attention to that same page 12—as to elimination of the variation in separate cylinder knocking tendency? A. I said in the next sentence: "The next most desirable result would be obtained by using a lead alkyl so volatile that at all times it would be in the vapor portion of the fuel charge and so be equally distributed among the cylinders. Such a lead alkyl does not exist."

Q. Which, if any, of the five lead alkyls we have been focusing our attention on most nearly approaches the theoretical highly volatile lead alkyl which you there men-
117 mentioned? A. Lead tetramethyl.

Q. If you used lead tetramethyl under the conditions that cause variation in separate cylinder knocking tendency, would its use eliminate that variation? A. No, it would not. Under some conditions it would leave the variation in separate cylinder knocking tendency substantially unaltered; and, as I stated a while ago, under other conditions it would result in a slight decrease of variation in separate cylinder knocking tendency.

Q. But suppose you used any one of the four other lead alkyls you have been discussing either alone or in combination of one, two, three, or all four of them. Would you reduce the variation in separate cylinder knocking tendency when conditions were such that there was such variation? A. Well, trimethyl ethyl——

Q. Do you mean alone? A. Alone, yes.

(Continuing)—under a limited range of operation with poor distribution would leave the variation unchanged. As I previously indicated, any one of the remaining three of the five would increase the variation in separate cylinder knocking tendency.

All of the mixtures which I described in my application increase the variation in separate cylinder knocking tendency.

Q. That is, to summarize, as I understand your testimony, when your invention as described and claimed by you is employed, you not only do not eliminate the variation of separate cylinder knocking tendency but you do not even reduce it? On the other hand, you increase it? Is 118 that correct? A. That is correct.

Q. Is there any further reference in your specification to the variation of separate cylinder knocking tendency? Here I will call your attention, to shorten the proceedings, to page 4.

Will you look at page 4 of your application? Will you read that brief description of Fig. 3 that you find there, and I will get the Fig. 3 chart out? Would you mind reading into the record what you find there? A. "Figure 3 is a chart showing, for conditions of imperfect fuel distribution, the variation in knocking tendency of these three cylinders of a multi-cylinder engine as the carburetor air-fuel ratio and per cent of fuel vaporization are varied."

Q. Will you please explain that chart, so that the Court may understand what it is all about? A. Figure 3 is a chart having for its vertical scale the highest useful compression ratio, or relative freedom from knocking tendency; and for its horizontal scale the percentage of fuel leaving the manifold in the vapor state.

Curves A, B, and C apply to cylinders A, B, and C which we discussed yesterday. In other words, curve A gives data for conditions of cylinder A, which, as we recall, was the lean cylinder receiving substantially only air and vaporized fuel.

Curve B is applicable to cylinder B, which received air, vaporized fuel, and a normal amount of liquid fuel; whereas curve C applies to cylinder C, which received air, vaporized fuel, a normal charge of liquid fuel, and in addition the

liquid fuel which under conditions of perfect
119 vaporization would have entered cylinder A.

It will be noted that curves A, B, and C were constructed using fuel containing no lead. It will be noted that when vaporization is imperfect there is a spread or a variation in the relative knocking tendency of the three cylinders. Cylinder A has the greatest tendency to knock, as indicated by its lowest H.U.C.R. Cylinder B has less tendency to knock, and cylinder C has the least tendency to knock.

To the same fuel was added lead in such proportions that the fuel in each cylinder, regardless of the amount of fuel in each cylinder, contained the same quantity of lead per gallon of fuel in that cylinder.

It will be noted that the addition of such quantity of lead had practically no effect on the variation in separate cylinder knocking tendency. Cylinder A now becomes A-prime. Cylinder B and cylinder C still have variation in separate cylinder knocking tendency, as they did before.

However, the conditions in all cylinders have been very greatly improved. Cylinder A, which had a very high knocking tendency relatively, without lead, has now moved up to a very much higher position, although the variation in separate cylinder knocking tendency remains.

It should perhaps be explained here that these curves were constructed, as I indicated——

Q. What do you mean by "these curves"? The record won't show that when you say "these curves." A. I am sorry. It should be explained that curves A-prime, B-prime, and C-prime were determined with fuels containing 3.15 grams of lead per gallon of fuel in each cylinder.

120 Q. In each instance? A. In each instance. That represents a better distribution of anti-knock material than I could obtain with any one of the mixtures which I have mentioned in my application. So, therefore, the use of any one of the mixtures which I have described would cause curves A-prime, B-prime, and C-prime to be separated further than they are in Figure 3.

Q. Let me interrupt for a moment. On page 12 of your application you say:

"The addition of the lead to the fuel changes neither the general shapes of the curves for the three cylinders nor the positions of the curves with respect to each other."

Will you include that in your explanation as you go along? A. Well, I think that is substantially what I have just indicated.

Q. I thought that was, but I wanted to be sure. A. Yes. Curves A-prime, B-prime, and C-prime have the same relative position that they did previously and substantially the same position with respect to each other.

Q. Have you mentioned, just to be sure, how those curves would look if you eliminated the variation in separate cylinder knocking tendency? A. If the variation in the separate cylinder knocking tendency were eliminated, curves A-prime, B-prime, and C-prime, would coincide on the curve for B-prime, which, as we recall, represents conditions existing in the normal cylinder. A-prime and C-prime would both move over into coincidation with B-prime.

121 Q. Just to make sure your explanation has included this point—if it has, please say so; if it has not, please add what you want to—you have stated that your mixture increases the variation in separate cylinder knocking tendency. Now, as to this Figure 3 of your application, you have stated that the addition of lead does not change the position of the curves with respect to each other. Have you already explained what apparent inconsistency there might be there? If not, will you please do so? A. Yes. I explained that when I indicated that curves A-prime, B-prime, and C-prime were determined under conditions where there was a constant concentration of lead—

Q. That is, you were feeding 3.15 grams to each cylinder in each instance, whereas in actual engine operation when variation does occur they would not be getting the same amount of grams of lead; is that correct? A. That is correct.

Mr. Wiegand. May I have the record show that the Figure 3 chart to which the witness has been referring is an enlarged photostat of Figure 3 of his application?

By Mr. Wiegand:

Q. Let us turn to page 5 of the Examiner's Statement, and that happens to be at page 68 of Plaintiff's Exhibit 1, in the lower right-hand corner. The Examiner says:

"However, applicant discloses the same composition as defined in the proposed claim and uses it in the same way."

Then, later down on the same page, he says, next to the last full paragraph:

"Thus it is noted that irrespective of the final
122 functional 'whereby' clause, the claim does define the
motor fuel composition disclosed by applicant and
therefore defines all that is material in applicant's invention."

What have you to say about those two statements? Consider first the first statement and then the second, and then both. A. Well, considering the first statement, "However, applicant discloses the same composition as defined in the proposed claim and uses it in the same way," I assume it is meant here that the "whereby" clause is a part of the claim.

Q. The first one is, "However, applicant discloses the same composition as defined in the proposed claim," and in the second one, he says, as I understand it, "Let us forget the 'whereby' clause."

The first one I want you to deal with as the Examiner put it, including the "whereby" clause as though it were in the claim as it is there. A. As I have said before, my object was to obtain the highest and most nearly uniform result over the wide range of operating conditions to be met in service, and that in doing so I have not eliminated the variation in separate cylinder knocking tendency. As a matter of fact, I have increased it. So I do not believe that the Examiner's suggested claim containing the "whereby" clause describes my invention at all.

Considering the second statement, "Thus it is noted that irrespective of the final functional 'whereby' clause, the claim does define the motor fuel composition disclosed by applicant and therefore defines all that is material
123 in applicant's invention," the claim without the "whereby" clause merely names the 5 lead alkyls—lead tetramethyl, lead tetraethyl, and the three intermediate ones—without giving any direction with respect to their use.

With such alkyls one would be free to make practically an infinite number of combinations of different percentages, 90 percent of which would be worse than bad tetraethyl. Only 10 per cent, or perhaps even less, would be better than lead tetraethyl.

Q. If unguided, it would take an infinite number of shots, unless you were exceedingly lucky, to find the 10 per cent of wheat out of the 90 per cent of chaff. Is that what you are trying to say? A. Yes. The chances would be about 10 to 1 against getting a combination, simply by random choice, which would be better than lead tetraethyl alone.

Q. And the lead tetraethyl, of course, we recall from yesterday, was the product that was on the market before you made your invention; is that correct? A. That is right.

Q. As I understand the testimony you have just been giving, speaking of that suggested claim, with or without the "whereby" clause the suggested claim does not define the motor fuel composition disclosed and claimed by you, does it? A. It does not.

Q. Now, I call your attention to some additional statements by the Examiner in the Examiner's Statement, paper No. 14, at the bottom of page 5 and the top of page 6. Your Honor, that is at page 68 of Exhibit 1. There the Examiner said:

124 "But considering applicant's argument more specifically that the claim is inoperative because it is not possible to 'eliminate the separate cylinder knocking tendency', it can only be pointed out that the dope comprising the mixed lead alkyls of different volatilities is an adjustment in this direction."

Then the Examiner says also, down below the middle of the page:

"Applicant's specification is a scientific treatise on the separate cylinder knocking tendency due to poor fuel distribution."

He goes on to say:

"This is the problem applicant solves and he does this by the use of a mixture of lead alkyls superior to solve this problem, because the separate cylinders get the anti-knock dope when they need it, notwithstanding none of the dope would be as good as lead tetraethyl per se in a perfect fuel distribution system."

Then he says:

"Thus the mixture of alkyls is most important to eliminate the separate cylinder knocking tendency, which varies with different operating conditions, while incidentally raising the anti-knock value of the fuel overall."

What have you to say about those four statements? A. Well, referring first to the statement at the bottom of page 5:

125 "But considering applicant's argument more specifically that the claim is inoperative because it is not possible to 'eliminate the separate cylinder knocking tendency', it can only be pointed out that the dope comprising the mixed lead alkyls of different volatilities is an adjustment in this direction,"

In the case of the mixtures which I have described there is not an adjustment in this direction. As I just pointed out, the use of any of my mixtures increases the variation in separate cylinder knocking tendency.

As to the Examiner's Statement, "Applicant's specification is a scientific treatise on the separate cylinder knocking tendency due to poor fuel distribution," it is true that the application does go in quite extensively to the treatment of the conditions arising because of poor fuel distribution. However—

Q. May I interrupt just a moment? Was your application based on pure theory alone or was it on the result of tests to which you have testified, as well as investigations? A. It was based entirely on investigation.

Q. Now, if you had not finished your answer, will you please proceed? A. At the bottom of page 6, quoting the Examiner's Statement again:

> "Thus the mixture of alkyls is most important to eliminate the separate cylinder knocking tendency, which varies with different operating conditions, while incidentally raising the anti-knock value of the fuel overall."

126 Well, that is diametrically opposite to the functioning of the mixtures which I describe. As I have said before, my object was to obtain an anti-knock agent which would give the highest and most nearly uniform result over the entire range of operating conditions and at the same time diminishing, insofar as possible, consistent with good overall operation, the knocking during conditions where poor distribution prevails. That is exactly opposite to the Examiner's interpretation of the object.

Q. And as to the Examiner's interpretation of what the invention is? A. Yes, that is correct.

Q. Now, I think that you have covered everything that I asked for except the third thing that I quoted, that is to say, on page 6 of the Examiner's Statement, where he said, "This is the problem applicant solves and he does this by use,'' and so forth. I need not read the rest of the sentence. You have it before you.

I do not think you included that in your answer. Will you discuss that? A. I did not re-read that, although it seems to me that what the Examiner says here is just another way of stating his interpretation. The Examiner states that the applicant solves this problem of poor distribution. I have indicated that—

Q. You have already answered? Is that what you mean? A. Yes, I think I have.

Mr. Wiegand. Does your Honor have the book of patents that the solicitor of the Commissioner handed up? There are four copies. I have an extra copy.

127 By Mr. Wiegand:

Q. Do you have that? A. Yes, I do.

Q. Now, referring to the Midgley Patent No. 1,573,846, which is in the Commissioner's answer, I ask you what kind of anti-knock compounds that patent refers to, and I direct your particular attention to the statements in that patent on page 2, second column, beginning at line 71, and then beginning at line 81. Do you have those places I mention? A. Yes.

Q. I ask you what kind of anti-knock compounds that patent refers to. A. This patent covers alkyl and phenyl compounds of lead, specifically mentioning ethyl, isopropyl, and methyl compounds. Also, mention is made of compounds of selenium, tellurium, tin, arsenic, and antimony, and the phenyl and alkyl derivatives of those metals.

Q. Will you turn to the Sullivan Patent No. 1,938,547? What in general is the object of that patent? A. Well, the object of that patent is the preparation of alkyl compounds of lead in the gasoline, as opposed to their manufacture in concentrated form and subsequent addition to gasoline.

Q. Look at the first paragraph and then look at page 2, line 81. Can you tell me what is there referred to? A. The first paragraph reads:

"This invention relates to processes of preparing hydrocarbon compounds of lead and it comprises a process wherein a hydrocarbon, for example hexane, is dis-
128 sociated by the action of an electric spark to liberate free alkyl radicals such as methyl, ethyl, propyl, or higher alkyl radicals, and these reaction products are caused to combine with lead whereby lead alkyl compounds are formed."

On the second page mention is made of the introduction into the process of methane, ethane, propane, or butane.

Q. Is the metal referred to in that patent chiefly lead? A. Yes.

Q. Take the Voorhees patent, No. 1,974,167. Will you discuss that rather briefly, not in too much detail? A. Well, the Voorhees patent has for its object fundamentally the object of the Sullivan patent.

Q. Refer to the bottom of column 1, page 1, of the Voorhees patent, and also to column 2, line 90, and then on page 3, column 2, beginning at line 97, where Voorhees states,

"Although I am not prepared to state the exact nature of the lead-hydrocarbon," and so forth. Will you explain those? A. Referring to page 1, column 1, line 47 or 48, there is this statement—and we should bear in mind that the object is to produce the compounds of lead in the gasoline—

"However, if desired, lead-hydrocarbon compounds having hydrocarbon radicals of the order of ethyl and propyl can be formed in the body of gasoline by dissolving therein, prior to chlorination, suitable quantities of ethane, propane, and butane, or unsaturated hydrocarbons," et cetera.

In the second column, line 90, there is a descrip-
129 tion of the operation of the process, in which this statement is made:

“Gasoline to be chlorinated enters the vessel by way of pipe 3 and provision for the introduction of ethane, propane, butane, or unsaturated hydrocarbons such as ethylene and propylene is made at pipe 4.”

On page 3, line 97, column 2, there is this statement:

“Although I am not prepared to state the exact nature of the lead-hydrocarbon compounds in my gasoline I believe that they are more complex in chemical structure than the simple alkyl compounds and their hydrocarbon radicals contain in general at least 4 carbon atoms. They are probably mixtures of lead derivatives of various isomeric liquid hydrocarbons having from 4 to 7 carbon atoms.”

Q. Now, in your application you state that you use lead alkyls having from 4 to 8 carbon atoms. Do you disclose on your charts in your application all of the lead alkyls within that range? A. No. In the application I stated that there are other lead alkyls in that range, but that inasmuch as they are not as effective as the lead alkyls in the ethyl-methyl series, they are omitted from the charts for reasons of simplicity.

Q. Now, take the Shappirio patent, No. 2,012,356. Can you tell us what compounds that patent is dealing with, and, to help you, look at page 1, column 2, in lines 47 to 49, and page 2, column 1, in the paragraph beginning on line 49, and then on the same page, 2nd column, in the paragraph starting at line 34, and then page 4, column 2, in the
paragraph beginning at line 13?
130 Have you those references? A. Yes, I have.

Q. Now, my question is, Will you tell us what compounds that patent is dealing with? A. It might be said, before the compounds are mentioned, that this patent also covers the manufacture of organo-metallic derivatives in the gasoline itself.

On page 1, column 2, line 9, there is this statement:

"It is desirable to have present some of the lower members of the paraffin series, and consequently gasoline containing such lower paraffins such as ethane, propane, and butane are particularly desirable for treatment in this process. Further gasolines containing aromatics are also highly desirable since these are readily converted into the desired metallo organic compounds * * *."

In column 2, line 46, there is this statement:

"Among the metals and non-metals that may be introduced into gasoline by the processes herein set forth there may be mentioned lead, tin, silicon, antimony, arsenic, phosphorus, mercury, thallium, etc."

On page 2, first column, the paragraph starting on line 49, there is this statement:

"As pointed out above, the process herein disclosed involves two essential steps, namely, first the treatment of the gasoline or analogous material to render it capable of being converted in part into metallo organic compounds, or the addition to the gasoline or analogous substance, of compounds that may be readily converted into such
131 metallic derivatives, and second the formation of the
metallo compounds therefrom. The metallic derivatives which it is desired to produce are the alkyls and aryls, such as lead tetra ethyl and antimony phenyl, and related substances."

On the same page, second column, line 34, there is this statement:

"Instead of proceeding in this way, the desired amount of a halogen derivative may be introduced into the gasoline. For example, ethyl chloride, methyl chloride, propyl bromide, etc., may be added to the gasoline or analogous material in the desired proportion to yield the requisite amount of metallo derivatives in subsequent operations."

On page 4, second column, line 14, appears this statement:

"Ordinarily as a result of this process, a complex mixture of organic derivatives of a stated metal, or metalloid is obtained, the derivatives, however, usually containing a number of closely related derivatives." They at this point are not named.

Q. Did anyone or all of these prior patents that you have just discussed teach or even suggest the combination recited in any of the claims in your application? A. No.

Q. Did they teach or suggest limiting the mixture to the five lead alkyls you are using in your application? A. No.

Q. Did these patents contain any guide of any
132 kind for choosing a mixture of lead alkyls to be used?

A. No. They are concerned entirely with the manufacture of organo metallic compounds, in some cases lead compounds only, without direction as to their use.

Q. Does that apply to Midgley as well, or did you mean the other three? A. I will exclude Midgley from that.

Q. That is, your last statement applied to Sullivan, Voorhees, and Shappirio? A. That is correct. Midgley does give directions for the use of his compounds—that is, particularly he gives directions with respect to the use of lead tetraethyl, but not the others.

Q. Then, as I understand it, the Sullivan, Voorhees, and Shappirio patents are directed to some special way of manufacture of these compounds in the gasoline itself, as distinguished from the usual way of making it separately and then adding it to the gasoline; is that correct? A. That is correct.

Q. Did any of these patents which you have been discussing disclose any recognition of the problem with which you were confronted and which you solved by making the invention which you are claiming? A. No, they make no mention of that problem.

Q. Does Midgley discuss anywhere in his patent the use of two or more of the lead alkyls at the same time
133 as proposed there? A. No, he does not.

Q. The Board of Appeals said in its decision that the patents which you have been discussing are suggestive of trying various selected mixtures of lead alkyls. Do you agree or disagree with that statement? A. I disagree with that.

Q. Will you please state why you disagree, briefly? I do not want you to get into too long a discussion. The statement of the Board of Appeals was to the effect that the patents which yu have just discussed are suggestive of trying various selected mixtures of lead alkyls. Do you disagree with it? A. I disagree with it. I do not know that any further explanation is needed. They simply do not make that suggestion.

Q. The patents do not? A. No.

Q. Let me put this question, and I think we had better have the chart of Figure 4 of the application again.

With that Figure 4 before you, does any of the patents you have discussed give any of the information as to the date represented by the curves E and F of your Figure 4? A. No.

Mr. Wiegand. That is all I have, your Honor.

The Court. We will suspend for ten minutes.

(A short recess was had, after which the following occurred:)

134 Cross-Examination

By Mr. Cochran:

Q. Mr. Bartholomew, I wish you would refer to the chart which is an enlargement of Figure 3 of your application and answer some questions concerning that.

I understand that the curves A, B, and C represent the highest useful compression ratio where there is no lead added to the fuel. That is right? A. That is correct.

Q. And that along a perpendicular line the difference in knocking tendency, for instance, between cylinder A and cylinder B, would be represented by the distance between the two at that point? A. That is right.

Q. So, in any case, for a given percentage of vapor in the fuel, the knocking tency would be measured by the vertical distance between the two curves? A. The difference in knocking tendency?

Q. The difference in knocking tendency, yes. A. Yes.

Q. Well, now, if you would take an engine in which the compression ratio was, for instance, 6.1—that is represented, I believe, on the vertical scale— A. That is right.

Q. Then, with no lead in the fuel and with, say, about an 80 percentage of vapor in the fuel, you would get a very great deal of knocking, would you not, in the A, B, and C cylinders? A. Yes.

Q. On the other hand, if you put in some of the
135 lead compounds, lead alkyls, as shown here, 3.15
grams per gallon, and used that engine with the same compression, you would not get any knocking in any of those cylinders, would you? A. That is right; all of them would have the knock eliminated.

Q. There would be no knocking in any of them anywhere along this range, say, down from 70 to 100 percentage of fuel vapor? A. That is correct.

Q. The knocking and the tendency to knock would be eliminated, would it not? A. That is right.

Q. Let us consider the other three curves, A prime, B prime, and C prime, of your Figure 3 in connection with the lower curves, A, B, and C. I believe you said that the difference in tendency to knock is measured by the distance between corresponding vertical points on the curves? A. Yes.

Q. Let us take points on the curves corresponding to about 78 per cent of fuel in the vapor state. Is it not true that the distance between the point on the curve C and the point on the curve A is much greater when there is no lead in it than when you have put lead in it? A. There is some reduction in the height of the line.

Q. Is it not a very distinct and material reduction? A. Well, I don't know. Yes, about 10 per cent, maybe; something like that.

Q. Is it not more like 25 per cent? A. Well, perhaps it
is. There are about 3½ divisions above—3.4, say; about
3½. And below there are, oh, 4.1, perhaps; some-
136 thing like that. There are perhaps 12 or 15 per cent;
something like that.

That represents a reduction, does it not, in the variation of tendency to knock when you put in the lead as compared to the tendency to knock when you do not have the lead? A. That is right, for the conditions existing when those particular tests were made.

Q. Yes. Well, is not that the reverse of what you said in your other testimony: that you increased, by putting in your compound, the difference in the tendency to knock? A. I do, except that I did not have my compound in the gasoline when this particular chart was made.

Q. This chart does not, then, prove anything about the matter? A. Well, yes it does, because my mixture is not distributed as well or in such a manner as to give the lead as good lead distribution among the cylinders as for this chart. The chart had to be drawn on some basis for purposes of illustration. I have constructed many other charts covering mixes which I have described.

Q. This chart does not prove your point; is that it? A. It does not prove the point except in this manner: that I have shown what happens with the concentration of lead indicated there. I have indicated in the specification that I am not able to obtain such an efficient distribution of lead with any of my mixtures. Therefore, my mixtures are worse with respect to distribution and also with respect to the effect on variations of cylinder knocking tendency.

Q. If we went by this chart, it would be contrary to your testimony as to elimination of variation of cylinder

137 knocking, would it not? A. Well, even that chart does not eliminate it. Within small limits there is no difference between the two.

Q. It decreases it, does it not, anyhow, as shown here? A. As shown there, there is a small decrease for that particular condition.

Q. This chart, then, does not demonstrate that you have proceeded in the opposite direction, does it? A. I do not say that it demonstrates anything except what the conditions thereon state—that is, except with respect to the conditions thereon stated.

Q. Well, taking these sets of curves anywhere from, say, 78 per cent fuel vapor on down to 100%, the variation of cylinder knocking as shown on this chart is always less when your products are added than it is without the products; is that right? A. Only when lead in that concentration was added. In my mixture—

Q. But this does not represent your mixture. A. That is right.

Q. These upper primes, A prime, B prime, and C prime, do not represent what you claim in your case? A. That is correct.

Q. Mr. Bartholomew, you do not always use the same mixture of these lead alkyls, do you? Is it not true that it is a matter of judgment, depending on the climate and the time of year? You do not have a definite percentage of tri-methyl-ethyl or di-methyl di-ethyl? A. No. As I state in the application, the proportions may be varied to suit the changes in climatic conditions or types of engine op-
138 eration or other factors that may have a bearing on the requirements.

Q. How do you determine that? By experiment? A. I have determined that by reference to Figure 4.

Mr. Wiegand. What was that question?

By Mr. Cochran:

Q. How do you determine what proportions of the ingredients to put in? Is it done by experiment? A. By a limited amount of experimental work after reference to the curves D, E, and F, and similar curves, which I said yesterday lie in between D and E but which are not shown on Figure 4, in the interest of simplicity.

Q. The materials you add to the gasoline to stop the knocking are the same materials as those specified in the claim as suggested to you by the Examiner? A. They are included among those named in the claim. Sometimes I use only three, sometimes four. In some cases it may be five, but not invariably.

Q. But all of those are included in the claim which was suggested to you by the Examiner? A. They are.

Q. Your objection to making the claim was that you did not think that those materials would produce the result which the Examiner said they would?

Mr. Wiegand. I do not know whether he was the one who decided to make the claim or not, but I am willing to have the answer come. That is a lawyer's problem.

Mr. Cochran. I will withdraw that question.

Mr. Wiegand. Go ahead with it if you want it.

The Witness. What was the question, please?

139 The Reporter (reading):

"Question. Your objection to making the claim was that you did not think that those materials would produce the result which the Examiner said they would?"

The Court. That is, the suggested claim?

Mr. Wiegand. Yes.

The Witness. My use of the materials does not produce the result outlined in the Examiner's suggested claim.

By Mr. Cochran:

Q. The materials are the same, are they not? A. Well, the materials used, irrespective of proportions, are the same materials. That is, I may or may not use all of those suggested by the Examiner.

Q. Well, if anyone used the same materials in the same proportions, he would get the same result? No matter what the Examiner said, he would get the same result? A. The same result?

Q. Regardless of what the Examiner thought the result might be or regardless of what the claim said the result would be, the materials would be the same, and the result would be the same? A. I take it that you mean that if the five materials suggested by the Examiner were used in a given proportion, the result from the same conditions would be always the same?

Q. Yes. A. That is correct.

Q. There are many of your claims which do not state any proportions; is not that true? A. In all cases they give direction.

Q. I should like you to refer to claim 6 of your
140 application.

Mr. Wiegand. May I show the witness the copy in the bill of complaint, so that we will have no confusion about it?

By Mr. Cochran:

Q. Considering, for example, claim 6, that claim represents:

> "A product comprising a motor fuel and a primary anti-knock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl."

That does not contain any directions or proportions, does it?

A. In this respect: three of the five are chosen.

Q. All those were included, however, in the claim suggested by the Examiner? A. They were named in the claim.

Q. You on occasion do use the others mentioned also in the claim? A. Under conditions outlined in the application, but not in a wide range of proportions. For example, tetramethyl: I have stated that if it is to be used at all, it should be used in small percentage, not exceeding 15 per cent.

Q. I should like you to refer to the patents on which you were examined this morning, the Sullivan, Voorhees, Shappirio, and Midgley patents. I merely want to ask you some general questions on them. A. Yes.

Q. It is true, is it not, that all of them disclose the use
of lead alkyls in connection with gasoline? A. I
141 believe that is correct. Yes, I believe that some mention of lead alkyls is made in each one.

Q. And all in connection with the use of gasoline to minimize knocking? A. Yes.

Q. Lead alkyls is a general name for the class of materials which includes the one you used in your claim or application? A. That is right.

Mr. Cochran. I think that is all.

Redirect Examination

By Mr. Wiegand:

Q. Will you take Figure 2 of your application—we have an enlargement of that on this chart—and explain to the Court how the lead alkyls are distributed? A. It will be recalled that cylinder A in Figure 2 received substantially only vaporized fuel along with the air entering cylinder A; that cylinder B received vaporized fuel and air, and also liquid fuel in the same proportions as in the mixture leaving the carburetor; that cylinder C received vaporized fuel, air, the normal amount of liquid fuel, and the liquid fuel which under conditions of perfect distribution would have entered cylinder A.

Now, the distribution of the lead alkyls follows the same pattern as the distribution of the fractions of the fuel. That is, vaporized lead alkyls is distributed substantially uniformly to all cylinders. Unvaporized lead alkyls are distributed unevenly among the cylinders. Cylinder C would receive the same excess of unvaporized lead alkyl as cylinder C receives the excess of unvaporized heavy ends of fuel.

* * * * * * * * * *

EXHIBITS.

157 *Petition*

To the Commissioner of Patents:

Your petitioner, Earl Bartholomew, a citizen of the United States and resident of Birmingham, County of Oakland, and State of Michigan, whose post-office address is P. O. Box 271, Birmingham, Michigan, prays that Letters Patent may be granted to him for the improvements in

—Method and Means for Improving Engine Operation—

set forth in the annexed specification.

And he hereby appoints Joe W. Morrison, c/o Ethyl Gasoline Corporation, Chrysler Building, New York City, Registration No. 11,083, his attorney, with full power of association, substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign his name to the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

EARL BARTHOLOMEW

Specification

To All Whom It May Concern:

Be it known that I, Earl Bartholomew, a citizens of the United States and resident of Birmingham, County of Oakland, and State of Michigan have invented certain new and useful improvements in

—Method and Means for Improving Engine Operation—

of which the following is a full, clear, concise and exact description, such as will enable others skilled in the art to which the invention relates to make and use the same.

158 This invention relates to modes for improving the operation of internal combustion engines. When lead tetraethyl was first used as an anti-knock agent, automobile engines were of the low duty type, i. e., the speed

range was relatively small, manifolds had relatively small passages and substantial heating of intake manifolds was commonly employed. Under these conditions, lead tetraethyl was one of the most efficient lead anti-knock compounds known and although other lead compounds, particularly lead alkyl compounds, were generally known and some tried, they did not possess the commercial practicability of lead tetraethyl.

It has been commonly understood that knocking in a given engine is the result of abnormal combustion, that its occurrence and severity under any given engine operating condition are governed chiefly, in the absence of added anti-knock compounds, by the chemical composition of the fuel, and that an improvement in conditions involving knocking revolves chiefly around a study of and changes in the base fuel. Gasolines are mixtures of hydrocarbons having a wide range of boiling points and molecular structure. The usual commercial gasolines have low-boiling fractions termed "light ends" which have good anti-knock value and higher-boiling fractions termed "heavy ends" which have relatively poor anti-knock value.

It is commonly known that knocking in automobile engines may occur at either high speed or low speed and that a given engine may knock at both high and low speeds. I have found that variations in conditions of operation within an engine, while other factors remain constant, have a greater influence on low speed knocking than the relative anti-knock values of the different fuel fractions. I
159 have also found that the "heavy ends", of lower anti-knock value, usually have little influence on the occurrence and severity of low speed knocking. Under conditions of high speed knocking, the "heavy ends", with the other fractions of the fuel, determine the over-all or unitary anti-knock value of a given fuel, but at low speed it is usually the lighter fractions, having the better anti-knock value, which control the occurrence and severity of knocking.

The characteristics of engines, base fuels and admixed anti-knock compounds are interdependent in the determination of the occurrence and severity of knocking. I have found that the different lead alkyl anti-knock compounds have certain different properties which, as related to certain properties of base fuels and features of engine design, form a broad background for improving engine operation. I have also found that these fundamental characteristics may be correlated to improve engine performance.

The objects of the present invention are to so direct this correlation that improved performance may be obtained under the varied conditions of operation of low duty and high duty internal combustion engines. By high duty engines I mean engines capable of operating at high volumetric efficiency over a wide range of rotative speeds.

I have found that mixtures of the three methyl-ethyl lead derivatives may be used, with or without other lead alkyls, with the various types of gasoline marketed to obtain better results in variable-speed, low duty and high duty engines than are obtainable under a wide range of operating conditions with any single lead alkyl. The three mixed methyl-ethyl lead derivatives are lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl. The problem of knocking is more difficult with the high duty engine and this invention finds its greatest utility in that field. The lead alkyls chosen for the lead alkyl mixture may be varied as well as their relative quantities to meet different conditions such as variations in the manner in which the car is driven, climatic conditions, the base fuel with which the anti-knock is used and the kind and condition of the engine. Where the engine may be operated with wide variance in some or all of these conditions the anti-knock mixture may be chosen in accordance with my invention so as to give a high average engine performance throughout.

In order that the invention may be more clearly understood, reference will now be made to the accompanying drawings, which illustrate engine operating conditions and also show the change of the anti-knock properties of the various lead alkyl compounds under varying engine operating conditions.

Figure 1 is a chart showing the octane number of that percentage of a representative commercial gasoline which is delivered in the vapor state by the manifold to the cylinders;

Figure 2 is a diagrammatic drawing, partly in section, of a portion of a six-cylinder engine with its intake manifold, carburetor and pistons. The pistons are all shown at their bottom dead center positions, giving equal volumes in all the cylinders, to permit the illustration of fuel distribution to three of the six cylinders at low engine speed when little or no heat is applied to the intake manifold;

Figure 3 is a chart showing, for conditions of imperfect fuel distribution, the variation in knocking tendency of these three cylinders of a multi-cylinder engine as the carburetor air-fuel ratio and per cent of fuel vaporization are varied;

Figure 4 is a chart with curves showing the trend of effectiveness of the lead alkyl anti-knock compounds, when added in equal concentrations of lead to a representative gasoline and utilized in the leanest cylinder of an engine operating at low speed with various percentages of fuel leaving the manifold in the vapor state. A similar curve is shown which is applicable to all the cylinders of the same engine under conditions of equal distribution of fuel to the cylinders.

161 I find that a better use of anti-knock agents depends primarily upon how the engine distributes and utilizes the gasoline-air mixtures. As gasoline enters the air stream in the carburetor of an internal combustion engine, the lightest or most volatile portions of the gasoline are vaporized and mixed with the air. The air-fuel vapor mixture and the unvaporized liquid fuel enter the intake manifold where the temperature is usually high enough to cause the vaporization of the more volatile portions of that part of the fuel which enters the manifold in the liquid state. The heavier portions of the fuel, except under con-

ditions of very high intake manifold temperatures seldom attained in the normal operation of automobile engines, remain in the liquid state until they enter the cylinders where they are normally completely vaporized before combustion starts. The percentage of the fuel vaporized in the manifold varies with the design of the engine, including the intake manifold, and with the conditions of engine operation.

Superimposed on the above described factors governing knocking under conditions of constant speed operation are those introduced by acceleration. When the throttle is opened suddenly, the greater inertia of the liquid fuel causes it to lag behind the air-vapor mixture in passing from the carburetor to the cylinders, with the result that missing would occur during the first few revolutions except for the additional fuel charge provided by the carburetor accelerating well or other means immediately after the throttle is opened. This additional fuel for acceleration provides additional light ends which, together with those in the constant speed charge, provide approximately maximum power mixtures in all the cylinders.

The fuel fractions or portions which are in the vapor state on leaving the manifold have varying octane numbers, depending on the composition of the fuel and the percentage of the fuel which is vaporized. Figure 1 illustrates how, for a representative fuel, the octane numbers of the vaporized fractions or portions leaving the manifold vary according to the extent of vaporization in the manifold and carburetor. 162

The term "octane number", as used herein, is intended to indicate the percent of 2.2.4 trimethyl pentane in a mixture of 2.2.4 trimethyl pentane and normal heptane which has the same knocking tendency as the fuel or fuel fraction in question when compared at their respective maximum knock air-fuel ratios at wide open throttle and low speed.

In the chart of Figure 1 the abscissa shows the percentage of a gasoline which is vaporized. The ordinate shows the octane number of the vaporized portion. That chart shows that when 55% of the fuel is vaporized the octane number of that 55% is about 67. When 80% of the fuel is vaporized the octane number of that 80% is about 58. The addition of the heavier ends by vaporization to the portion already vaporized markedly reduces the octane number of the resulting vapor portion. It follows that the smaller the vaporized portion of the fuel the higher is its octane number. The curve of Figue 1 is representative of practically all gasolines now marketed.

In Figure 2, three cylinders of a six-cylinder engine are shown diagrammatically, by way of example, to illustrate three different mixture conditions in the cylinders. In this Figure the engine 10, has a curburetor 11, a down-draft manifold 12, with its horizontal header 13, intake ports 14, exhaust ports 15 and pistons 16. The cylinders are lettered A, B and C. The pistons are not shown in their normal relative positions, connected to a crank-shaft. Instead, all are shown in their bottom dead-center positions to illustrate the equal capacity of the cylinders for receiving a charge and to illustrate the nature of the charges received by the cylinders when the engine is operating at wide open throttle and low speed.

Vapor-air mixtures are indicated in Figure 2 by the broken cross-hatch lines, the relative concentration of the vapor being indicated by the relative heaviness of the cross-hatch lines. Unvaporized heavy ends of the gasoline
163 are shown in the figure as drops 17, which are ultimately vaporized in the cylinder by heat from residual exhaust gas, metal surfaces and other sources. This showing is purely diagrammatic and not intended either to represent position or to be quantitative.

In the operation of an automobile engine the portion of the fuel usually vaporized in the carburetor and manifold (for example 55 to 90 percent) is distributed in substantially equal quantities to all of the cylinders with substantially equal quantities of air. In the distribution of the heavier portions of the fuel, not vaporized in the carburetor or manifold, there are three significant conditions met in a multi-cylinder engine, as follows:

(1) One or more cylinders A may receive only light ends vaporized in the carburetor and manifold, and substantially no liquid heavy ends.

(2) One or more cylinders B may receive vaporized light ends and liquid heavy ends in such proportions that the composition of the fuel and ratio of weight of air to weight of fuel are substantially the same as for the mixture leaving the carburetor.

(3) One or more cylinders C may receive vaporized light ends and liquid heavy ends in the same ratio as under conditions (2) referred to above, and, in addition, the liquid heavy ends which under conditions of perfect distribution would have entered the first mentioned cylinders A. These three conditions are the result of the uneven distribution of the heavier liquid portions of the fuel to the several cylinders, the distribution being influenced by the dimensions and shapes of the manifold, the speed of the engine, and other factors. The heavy ends which enter the cylinders as liquid under the latter two conditions may be vaporized immediately upon contacting the heated cylinder and piston surfaces but in any case are usually vaporized before combustion is initiated. The cylinders receive substantially the same quantity of air but different quantities
and compositions of fuel. The richness of the mix-
164 ture or the relative quantity of fuel present is indi-
cated in Figure 2 by the heaviness of the cross-hatch
lines. Cylinder A is seen to have a large ratio of air to fuel, cylinder B a smaller ratio of air to fuel, and cylinder C a still smaller ratio of air to fuel. Cylinder A has the leanest mixture and cylinder C has the richest mixture. The additional fuel in cylinder C, which causes this mixture to be the richest, consists of the heavy ends which, under conditions of good distribution, would have entered cylinder A and which we see from Figure 1 have low anti-knock

value. Cylinder A which has the leanest mixture contains the part of the fuel having the best anti-knock value but, owing to the leanness of the mixture, the knocking tendency of the mixture in cylinder A is greater than that in cylinder B. The knocking tendency of the mixture in cylinder B, in turn, is greater than that in cylinder C, which has the richest mixture, as will be explained hereafter. These and intermediate conditions normally exist when low speed knocking occurs. I find that during conditions existing in the summer time the unequal distribution of fuel described above occurs at substantially constant car speeds up to about 25 miles an hour. Under more adverse conditions, such as winter operation, less heat applied to the intake manifold or the use of less volatile fuel, the unequal distribution of fuel may persist at even higher car speeds.

At substantially constant car speeds above approximately 35 miles an hour the velocity of the mixture of air and fuel vapor in the intake manifold is usually sufficiently high to entrain the liquid portion of the fuel and distribute it in substantially equal quantities to all the cylnders. Under this condition all the cylinders receive charges which are substantially the same and the charges in all the cylinders have the characteristics of the charge in cylinder B in Figure 2.

Equal distribution of the fuel at low speed also occurs if the intake manifold temperature is sufficiently high
165 to vaporize all the fuel. This condition may occasionally exist during hot weather operation. After a car has been driven at high speed during hot weather, the heat built up in the engine is sometimes sufficient to cause complete vaporization of the fuel in the intake manifold during low speed operation immediately following. The effects of increased temperature and of increased speed are to improve distribution, hence the range of engine speed over which good distribution occurs is governed by a combination of factors such as engine speed and load, climatic conditions and the type and condition of the engine itself.

Under conditions of wide-open throttle operation at constant low speed (for example when ascending a hill), the carburetor air-fuel ratios of automobile engines, now being marketed, are usually in the range from 10.0 to 1, to 12.5 to 1, and the mixture ratio of the leanest cylinder under the same conditions usually does not exceed about 14.0 to 1. Carburetor air-fuel ratios in this range are employed for the development of maximum engine power under conditions of poor distribution.

I find that under such conditions the various cylinders receive mixture ratios ranging between the leanest, which is usually about 14.0 to 1, and the richest, which is richer than the carburetor air-fuel ratio of 10.0 or 12.5 to 1. During manual or automatic choking in periods of warming up or very low atmospheric temperature, carburetor air-fuel ratios may be much richer than 10.0 to 1. They are usually richer also immediately after the quick opening of the throttle during acceleration.

Fuel octane number and mixture ratio are factors which influence the design of an engine for the production of maximum power and economy. The highest compression ratio that may be used without knocking, commonly called the highest useful compression ratio (H.U.C.R.), is a measure of the utility of these two factors when other factors remain constant. Figure 3 illustrates the H.U.C.R. of cylinders A, B and C of Figure 2 as carburetor air-fuel ratio and fuel vaporization in the carburetor and manifold are varied.

166 The chart of Figure 3 has as its scales of abscissae the carburetor air-fuel ratio and the per cent of vaporized fuel leaving the manifold. The chart has as its left scale of ordinates the highest useful compression ratio (H. U. C. R.). The chart has as its right hand scale of ordinates the air-fuel ratio of cylinder C. Curves A, B and C of the chart show the highest useful compression ratio of cylinders A, B and C of Figure 2 as the carburetor air-fuel ratio and per cent of vaporized fuel leaving the manifold

are varied. For purposes of comparison and because the mixture ratio in the leanest cylinders of automobile engines does not vary widely, I here choose to consider only those cases where the mixture ratio in the leanest cylinder (A) is constant and is of the order of 14.0 to 1. In order for cylinder A to operate at this constant air-fuel ratio as the percent of fuel vaporized in the carburetor and manifold decreases, it is necessary that the carburetor air-fuel ratio be increased. Cylinder B is assumed to receive fuel of the same composition and to have the same mixture ratio as that delivered by the carburetor. Cylinder C is assumed to receive the same fuel as received by cylinder B plus the liquid portions which under conditions of perfect distribution would have entered cylinder A. Cylinder C has the richest mixture and its mixture ratios are read from the right hand scale of ordinates.

Curve B, of Figure 3, shows that the highest useful compression ratio of cylinder B, of Figure 2, varies as the mixture ratio in the cylinder is varied. At a cylinder air-fuel ratio of about 13.8 to 1 the knocking tendency of the mixture in the cylinder is greatest, as is indicated on the curve by the lowest H. U. C. R. This air-fuel ratio at which the knocking tendency is greatest is known as the maximum knock air-fuel ratio. At mixture ratios richer or leaner than the maximum knock air-fuel ratio the knocking tendency of
the mixture in the cylinder is less and the diminution
167 in knocking tendency becomes greater as the mixture
ratio is made progressively richer or leaner.

Curve A shows that as the percentage of fuel vaporized in the carburetor and manifold decreases, and as the carburetor air-fuel ratio is made correspondingly richer to maintain the air-fuel ratio constant in cylinder A, the highest useful compression ratio for cylinder A also increases, notwithstanding the fact that its air-fuel ratio remains constant. This is due to the fact that the fuel entering cylinder A consists of progressively lighter ends which have correspondingly higher octane numbers. The resulting increases in H. U. C. R. of cylinder A due to the higher anti-knock value of these lighter ends is not as great as the corresponding increases in H. U. C. R. of cylinder B obtained by the accompanying richening of the mixture therein.

As the percentage of fuel vaporized in the carburetor and manifold is decreased, and the carburetor mixture is correspondingly richened, cylinder C receives an increasing proportion of heavy ends of low octane number, hence the octane number of the fuel in cylinder C decreases. However, curve C of Figure 3 shows that as the carburetor mixture is richened cylinder C knocks progressively less. This is due to the fact that as the mixture in the cylinder is richened, the resultant decrease in knocking tendency is more than sufficient to offset the effect of the lower octane number of the fuel. The result is that under engine operating conditions with poor distribution, the H.U.C.R. of cylinder C is greater than that for either cylinder B or cylinder A. The H.U.C.R.s of the several cylinders vary more because of changes in cylinder air-fuel ratio under conditions of imperfect distribution than they do because of changes in the relative anti-knock value of the light and heavy ends of the fuel.

Curves A′, B′ aud C′ of Figure 3 show the highest useful
compression ratio for cylinders A, B and C respec-
168 tively when the fuels of curves A′, B′ and C′ contain
a lead alkyl in a concentration of 3.00 grams of lead
per gallon of fuel received by the cylinder. The addition of the lead to the fuel changes neither the general shapes of the curves for the three cylinders nor the positions of the curves with respect to each other. Rich cylinder C without lead in the fuel has a higher useful compression ratio than lean cylinder A with lead in the fuel when less than approximately 75 % of the fuel leaves the manifold in the vapor state.

Cylinder A, containing substantially only the lighter ends of the gasoline, has the leanest mixture ratio and is in greatest need of the assistance of lead alkyl anti-knock compounds. In a multi-cylinder engine there may be several cylinders having the conditions of cylinder A and these conditions shift around among the cylinders during operation. The ideal solution of this problem would be to distribute the anti-knock agent in accordance with the needs of each cylinder but this is impractical. The next most desirable result would be obtained by using a lead alkyl so volatile that at all times it would be in the vapor portion of the fuel charge and so be equally distributed among the cylinders. Such a lead alkyl does not exist.

Other important factors involved, in addition to the factor of lead alkyl distribution to the cylinders, are the inherent anti-knock effect of the different lead alkyls (anti-knock effect per unit weight of lead) and the variation in inherent anti-knock effect under different operating conditions. I find that the apparent advantage of low boiling lead alkyls during poor distribution is offset in varying degrees by the relatively low inherent anti-knock effect of these alkyls during good distribution. I also find there is an advantage in using a mixture containing certain low boiling lead alkyls and certain lead alkyls which have higher inherent anti-knock effect, and which have less variation in anti-knock effect as operating conditions change even though the latter lead alkyls have a higher boiling point.

The curves of Figure 4 show the octane numbers of
169 the vaporized portions of mixtures of the base fuel of
Figure 1 and some of the lead alkyls under three representative operating conditions of cylinder A, namely with 53% (poor distribution, curve E), 87% (good distribution, curve F), and 100% (perfect distribution, curve D) of the fuel leaving the manifold in the vapor state. The condition of 100% of the fuel leaving the manifold in the vapor state for cylinder A is also representative of the same condition for cylinders B and C. In Figure 4, one scale of abscissae denotes some of the lead alkyls by the number of carbon atoms in the lead alkyl molecules, up to and including 12 carbon atoms, and the other scale of abscissae the corre-

sponding boiling points of these alkyls at a pressure of 13 millimeters of mercury. The scale of ordinates shows octane numbers. In all cases the base fuel leaving the carburetor contains a single lead alkyl in a concentration of 3.00 grams of lead per United States gallon.

Curve D on the chart shows the octane number of the mixture of fuel and anti-knock compound in all the cylinders of an engine operating at low speed with perfect distribution. Under these conditions the concentration of anti-knock compound in the fuel and the composition of the fuel in each of the cylinders is the same as in the fuel leaving the carburetor. This curve shows the relative efficiency of the lead alkyls at low engine speed when no distribution problem exists, and will hereafter be referred to as the curve of inherent anti-knock value at low speed. As such it serves as a reference curve. I have found that lead alkyl anti-knock compounds containing both methyl and normal propyl radicals, such as lead tri-methyl propyl, and compounds containing iso-propyl radicals, such as lead di-ethyl diisopropyl, have lower inherent anti-knock effect than compounds shown on curve D of Figure 4. Since those compounds having lower inherent anti-knock effect have less commercial utility the chart of Figure 4 has been simplified by omitting them.

170 Lead tri-ethyl methyl has the highest inherent anti-knock value under the low speed conditions shown by curve D, and the inherent anti-knock effect of the other lead alkyls diminishes as the number of carbon atoms is increased or decreased. Of the alkyls shown on curve D, lead tetramethyl has the lowest inherent anti-knock value. I find that in some commercial gasolines the 7 carbon atom compound (lead tri-ethyl methyl) is a little better than the 8 carbon atom compound (lead tetra-ethyl), and in other commercial gasolines the 8 carbon atom compound is better than the 7 carbon atom compound. The difference between the two in any case is small, and so I consider them as on a par.

Curve E shows the relative anti-knock effect of the same lead alkyls under low speed condition where only 53% of the fuel is delivered in the vapor state by the manifold to cylinder A of Figure 2. Lead tetramethyl shows the greatest anti-knock effect, and the effectiveness of the other alkyls decreases progressively with an increase in the number of carbon atoms in the molecule. The greater anti-knock effect of the alkyls having a small number of carbon atoms is due to the higher volatility of these compounds which results in a greater percentage being distributed with the portion of the fuel in the vapor state. Curve F is a similar curve for engine operating conditions where 87% of the fuel is delivered in the vapor state to cylinder A of Figure 2. It will be noted from these three curves that, as engine conditions are changed toward perfect fuel distribution, the effectiveness of lead tetramethyl and lead tri-methyl ethyl falls toward their points on the inherent curve D while the effectiveness of the remaining alkyls rises toward their points on the inherent curve D.

The high effectiveness of the low carbon atom lead alkyls is found in slow speed winter time driving before the engine is warmed up. As the engine warms up the operating conditions approach those given on curve D. The high apparent effect of lead tetramethyl may be useful for but
171 a short period of driving. The problem of the distribution and utility of the lead alkyls is emphasized during engine acceleration. During the first few revolutions after the quick opening of the throttle, all the cylinders receive substantially only vaporized fuel, and therefore lead alkyls are effective during this period insofar as they are vaporized and enter the cylinders with the vaporized fuel. Good volatility of the alkyl is desirable to meet this condition.

At higher engine speeds or higher manifold temperatures where substantially uniform distribution of fuel occurs, the anti-knock agent is distributed substantially equally and inherent anti-knock effectiveness is the controlling factor because at high engine speeds engines are usually quite sensitive to small changes in the anti-knock value of the fuel. At high car speed under conditions of perfect distribution the lead alkyls containing from 8 to 12 carbon atoms have substantially the inherent anti-knock values shown on curve D. The 6 and 7 carbon atom compounds have slightly less anti-knock value than the 8 carbon atom compound and the 4 and 5 carbon atom compounds considerably less inherent effectiveness than indicated by curve D, lead tetramethyl having the greatest loss in inherent effectiveness as engine speed is increased. Because of their greater loss of inherent anti-knock value at high car speed, lead tetramethyl and lead tri-methyl ethyl do not have as high overall effectiveness relative to lead di-methyl di-ethyl and lead tri-ethyl methyl as indicated by the points representing these compounds on Curve D. The boiling points of the compounds containing 6 or more carbon atoms, are a deterrent to their use under conditions of poor distribution. I have found that the boiling points of the lead alkyls are correlated with engine operating conditions instead of with the boiling points of the various fractions of the fuels.

The addition of an anti-knock agent to a base fuel increases the H.U.C.R. of an engine using this fuel. I find
that the best anti-knock agent is a mixture of lead
172 alkyls which gives the highest and most nearly uni-
form increase in H.U.C.R. of an engine over the
range of operating conditions under which it is used. I find that this mixture should contain in substantial quantity a lead alkyl which has high inherent anti-knock effectiveness over a wide range of engine speeds and as low a boiling point as possible consistent with this requirement, and that the 7 carbon atom compound, lead tri-ethyl-methyl, best meets these conditions. I find that this mixture should also contain in substantial quantity a lead alkyl which has a low boiling point, relatively high effectiveness under conditions of poor distribution and not too low inherent anti-knock effectiveness over a wide range of engine speeds, and that the

5 carbon atom compound, lead tri-methyl-ethyl, best meets these conditions. The 6 carbon atom compound, lead di-methyl di-ethyl, has a relatively low boiling point and a relatively high and substantially uniform anti-knock effectiveness over a wide range of engine speeds and I find that this compound should be used in the mixture in substantial, or even predominant, proportions. To meet the widest normal range of operating conditions my preferred mixture consists, on a volume basis, of 25 percent lead tri-methyl-ethyl, 45 percent lead di-methyl di-ethyl and 30 percent lead tri-ethyl methyl. With this mixture I use one or more compounds, such as organic halides, in accordance with and for the purposes stated in Midgley United States Patent No. 1,592,954, issued on July 20, 1926 and entitled "Fuels". I prefer to use, on a volume basis, in the final mixture, 23 percent of ethylene di-bromide, 14 percent of ethylene di-chloride and 63 percent of the lead alkyl mixture.

The quantity of such mixture that is added to fuels depends on the composition of the fuels and the desired improvement in the anti-knock value of the fuels. To one United States gallon of an average commercial gasoline as marketed in the United States, I usually add up to about 4.5 c.c. of the final mixture; however, larger concentrations may be used.

173 In the practice of my invention, a fuel-air mixture is compressed, in an internal combustion engine, to a compression pressure greater than the critical compression pressure of the base fuel, and the fuel-air mixture is burned with the mixture of lead alkyls, with a resultant decrease in knocking tendency. The final mixture containing lead alkyls may be mixed with the base fuel to form a new treated fuel or it may be injected into the intake manifold. In the former case the final mixture is carbureted and distributed with the fuel-air mixture.

Lead tetramethyl and lead tetra-ethyl may be used in the mixture and when these compounds are used my preferred mixture consists, on a volume basis, of 4.0 per cent lead tetramethyl, 23 percent lead tri-methyl ethyl, 40 percent lead di-methyl di-ethyl, 27 percent lead tri-ethyl-methyl and 6 percent lead tetra-ethyl. The proportions of the methyl-ethyl lead alkyls, and of the tetra-methyl and tetra-ethyl compounds if the latter are used, may be varied to meet different ranges of climatic conditions, operating conditions and engine design. When cold weather or relatively low speed driving conditions prevail, a larger proportion of the lower boiling compounds and a smaller proportion of the higher boiling lead alkyl compounds may be used. If lead tetra-methyl is used in the mixture, I prefer to keep its concentration low and even in the coldest weather to have its concentration less than 15 percent. When hot weather or relatively high speed driving conditions prevail, a higher proportion of the higher boiling compounds, and particularly lead tetra-ethyl, and a smaller proportion of the lower boiling compounds may be used. The exact proportions to be used depend on the specific conditions to be met.

Other lead alkyls may be used in the mixture but if present I prefer that they be employed in relatively small concentrations. The organic halides may be used with all these mixtures.

The primary or basic anti-knock agent is the mixture of the three methyl-ethyl lead derivatives—lead tri-methyl ethyl, lead di-methyl di-ethyl and lead tri-ethyl methyl.

This primary or basic agent may be used alone or
174 with other anti-knock compounds which modify the
anti-knock effectiveness provided by different mixtures of these three compounds. Where tetra-methyl lead is added I prefer that its concentration should not exceed 15 percent based on lead present.

As stated above lead tri-ethyl methyl and lead tetra-ethyl are about on a par except for boiling point and the difference in the boiling points of these two compounds is not so material in hot weather. For these reasons lead tetra-ethyl is taken as an equivalent for lead tri-ethyl-methyl in an anti-knock mixture for hot weather, and such mixture may have a relatively large concentration of either or both of these compounds. Except for this hot weather mixture I prefer the concentration of other anti-knock compounds to be relatively small so as to merely modify the properties of the primary or basic anti-knock agent.

I have found that when using the mixture or mixtures described above, under the conditions of imperfect fuel distribution described with reference to cylinders A, B and C, that the knocking of the cylinder A is reduced to a minimum consistent with good engine performance over the entire range of operating conditions.

The problems of distribution and of the use of lead alkyls, as anti-knocks, have been applied to a base fuel of the type shown in Figure 1. In the case of base fuels having a substantially flat characteristic curve, i.e., all fractions of approximately equal octane number, the H.U.C.R. of cylinder A of Figure 2 remains constant as the carburetor air-fuel ratio is varied and causes curve A of Figure 3 to become a substantially straight, horizontal line. As the carburetor air-fuel ratio is made richer than that for maximum knock, the H.U.C.R. of cylinders B and C increases faster than in the case of the fuel of Figure 1 since there is no depressing effect of heavy ends of low anti-knock value. Therefore cylinder A, relative to cylinders B and C, is in even greater need of the assistance of lead alkyl anti-knock compounds.

When the base fuel has a characteristic curve with
175 with a slope opposite to that of the curve of Figure
1, i.e., having heavy ends with higher anti-knock value than the lighter ends, the H.U.C.R. of cylinder A of Figure 2 decreases as the carburetor air-fuel ratio is made richer than that for maximum knock, and the H.U.C.R. of cylinders B and C increases more rapidly than in either previous case as the heavy ends of high anti-knock value are added. Cylinder A, therefore, has the greatest possible need of the assistance of lead alkyls. I find that the use of fuels of the last two types described does not materially alter the problem of the selection of the best lead alkyl mixture.

Curve D of Figure 4 is applicable to base fuel stocks which constitute by far the major portion of the gasoline sold in the United States. A few unusual gasolines have curves of inherent lead alkyl anti-knock value which differ from curve D of Figure 4 in that the compound of maximum effectiveness is other than that containing seven or eight carbon atoms. However, in these fuels I find that the above described preferred mixtures give the best over-all results.

My invention finds its greatest utility in high-duty engines. It is also useful in low-duty engines, particularly in the winter-time or under variable conditions of operation.

It will be appreciated from the above description that the invention not only resides in the method of operating a gasoline engine by means of the combustion of gasoline with the aforesaid anti-knock mixture or mixtures, but also resides in the various anti-knock mixtures as hereinbefore described, either alone or admixed with gasoline.

It will also be appreciated that the invention is not only applicable for use with gasoline, such as the ordinary gasoline of commerce, but is also applicable for use with mixtures of gasoline and other fuels, for instance, alcohol or benzene-gasoline mixtures.

176 I claim:

1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

2. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a

primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl in which antiknock mixture the lead dimethyl diethyl predominates.

3. The method of operating an internal combustion engine
which comprises forming a combustible mixture of a motor
fuel and air, compressing the mixture to a compression
pressure greater than the critical compression pressure of
the motor fuel and burning the mixture together with a
primary antiknock agent comprising a mixture of lead tri-
methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-
methyl, and lead tetraethyl the concentration of the
177 lead being greatest in the lead triethyl methyl and
lead tetraethyl.

4. The method of operating an internal combustion engine which comprises forming a cumbustable mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of the three methyl-ethyl lead derivatives; and lead tetramethyl in a concentration not exceeding fifteen percent.

5. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of the three methyl-ethyl lead derivatives.

6. A product comprising a motor fuel and a primary antiknock agent consisting of the three methyl-ethyl lead derivatives.

7. A product comprising a motor fuel and an anti-knock
material having as its primary antiknock com-
178 ponent a mixture of the three methyl-ethyl lead de-
rivatives, in which mixture the concentration of
lead dimethyl diethyl predominates.

8. A product comprising a motor fuel and a primary antiknock agent consisting of the three methyl-ethyl lead derivatives, in which the concentration of lead dimethyl diethyl predominates.

9. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of the three methyl-ethyl lead derivatives, and lead tetramethyl in a concentration not exceeding fifteen percent.

10. A product comprising a motor fuel and a primary antiknock agent consisting of the three methyl-ethyl lead derivatives, and lead tetramethyl in a concentration not exceeding fifteen percent.

11. A product comprising a motor fuel and an antiknock agent comprising lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl and in which the greatest lead concentration is in the group consisting of lead triethyl methyl and lead tetraethyl.

179 12. A product comprising a mixture of a motor fuel, a primary antiknock agent consisting of the three methyl-ethyl lead derivatives; and lead tetramethyl and lead tetraethyl present in relatively small concentrations.

13. A product comprising a mixture of the three methyl-ethyl lead derivatives and an organic halide.

14. A product comprising a mixture of the three methyl-ethyl lead derivatives, at least one other lead alkyl present in a relatively small proportional quantity, and an organic halide.

15. A product comprising a mixture of the three methyl-ethyl lead derivatives and lead tetramethyl in a concentration not exceeding fifteen percent.

16. A product comprising a mixture of lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl with the greatest lead concentration in the lead triethyl methyl and lead tetraethyl.

180 17. A product comprising a mixture of the three methyl-ethyl lead derivatives, lead tetramethyl in a concentration not exceeding fifteen percent and an organic halide.

18. A product comprising a mixture of lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl with the greatest lead concentration in the lead triethyl methyl and lead tetraethyl, and an organic halide.

19. The improved antiknock agent; and product comprising a motor fuel and improved antiknock agent; and method of operating an internal combustion engine substantially as described and for the purpose specified.

181 In Testimony Whereof I affix my signature.

EARL BARTHOLOMEW.

Oath

State of Michigan, County of Wayne, ss:

Earl Bartholomew, the above named petitioner, being duly sworn, deposes and says: that he is a citizen of the United States and a resident of Birmingham, County of Oakland, and State of Michigan, that he verily believes himself to be the original, first and sole inventor of the improvement in

—Method and Means for Improving Engine Operation—

described and claimed in the annexed specification; that he does not know and does not believe that the same was ever known or used before his invention or discovery thereof, or patented or described in any printed publication in any country before his invention or discovery thereof, or more than two years prior to this application, or in public use or on sale in the United States for more than two years prior to this application; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives or assigns more than twelve months prior to this application; and that no application for patent for the said invention has been filed by him or his legal representatives or assigns in any country foreign to the United States.

EARL BARTHOLOMEW.

Sworn to and subscribed before me this 1st day of September 1936.

MARIE M. MARTIN,

(Seal) *Notary Public, Wayne Co.,*

My Commission expires Oct. 14, 1936

FIG.1.

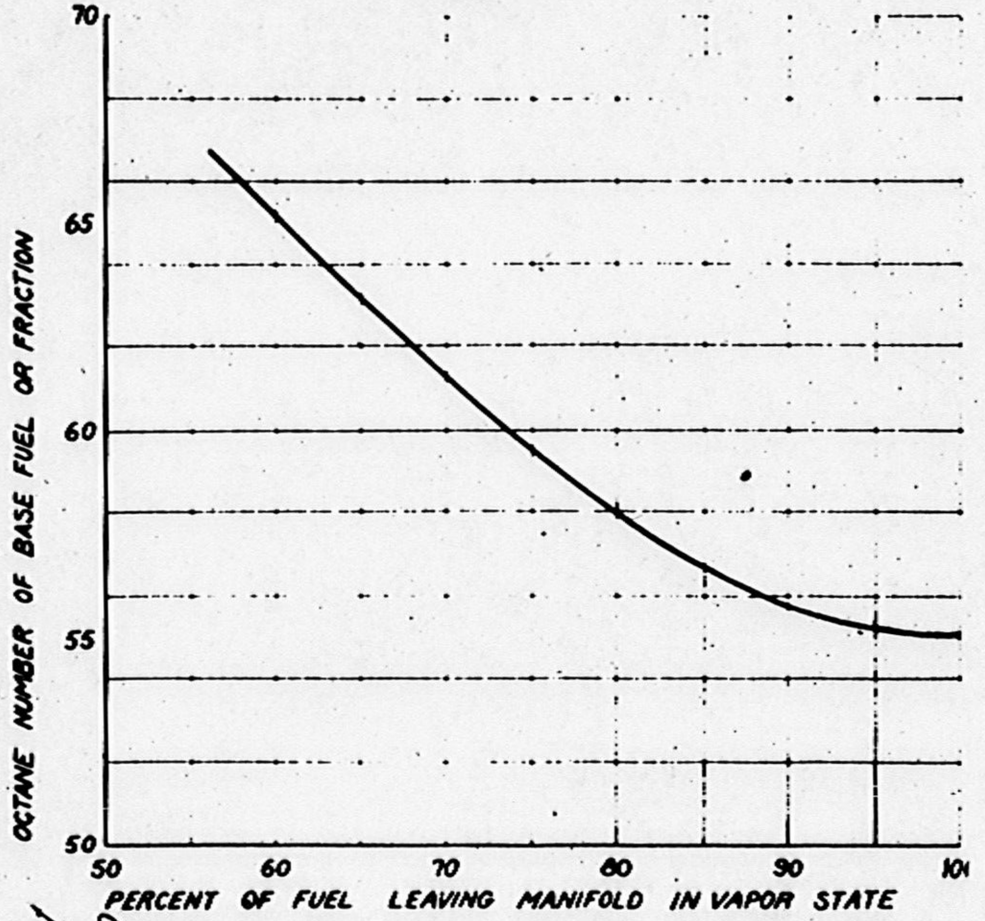
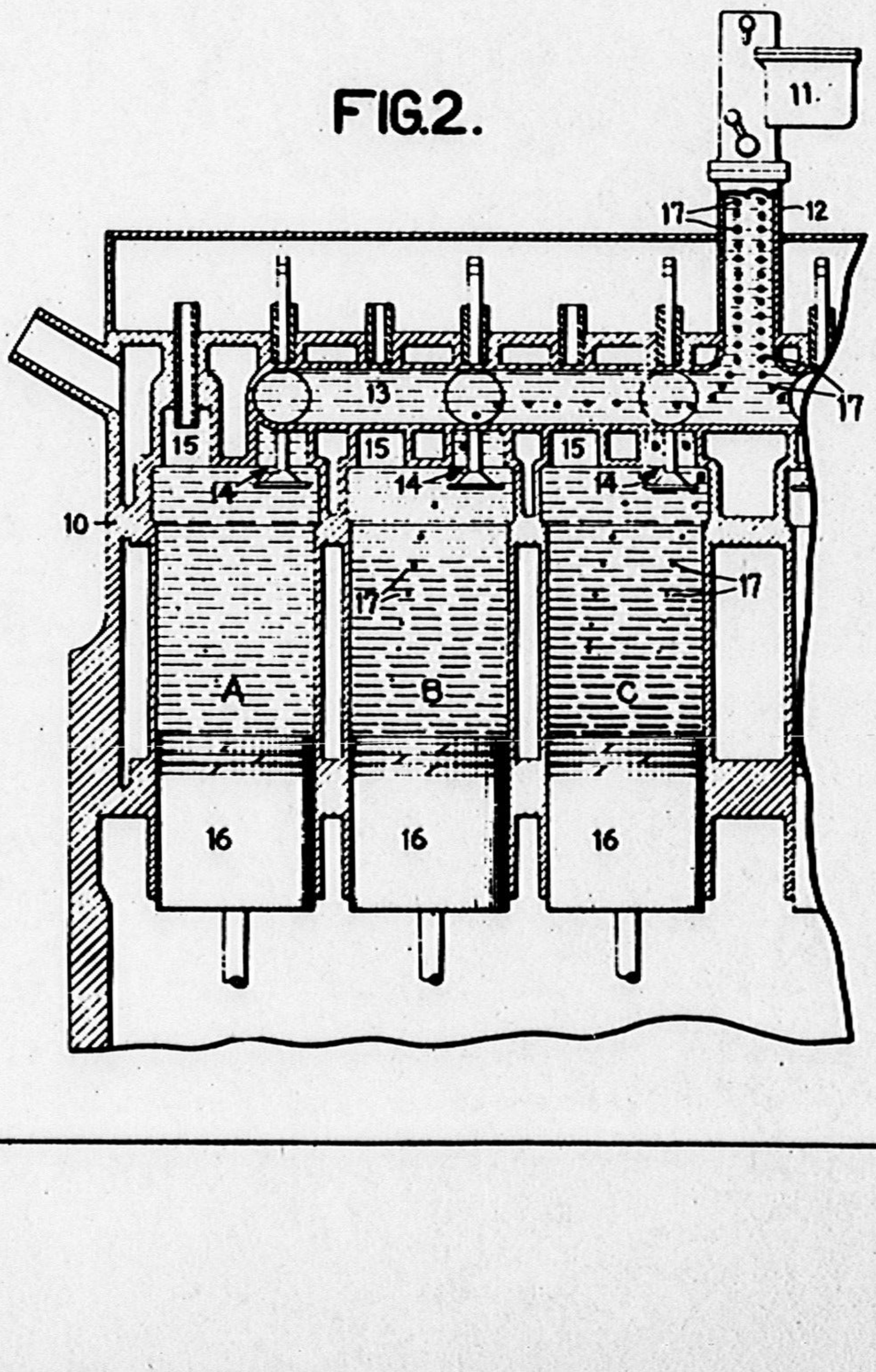


FIG.2.



INVENTOR
Earl Bartholomew
BY

ATTORNEY

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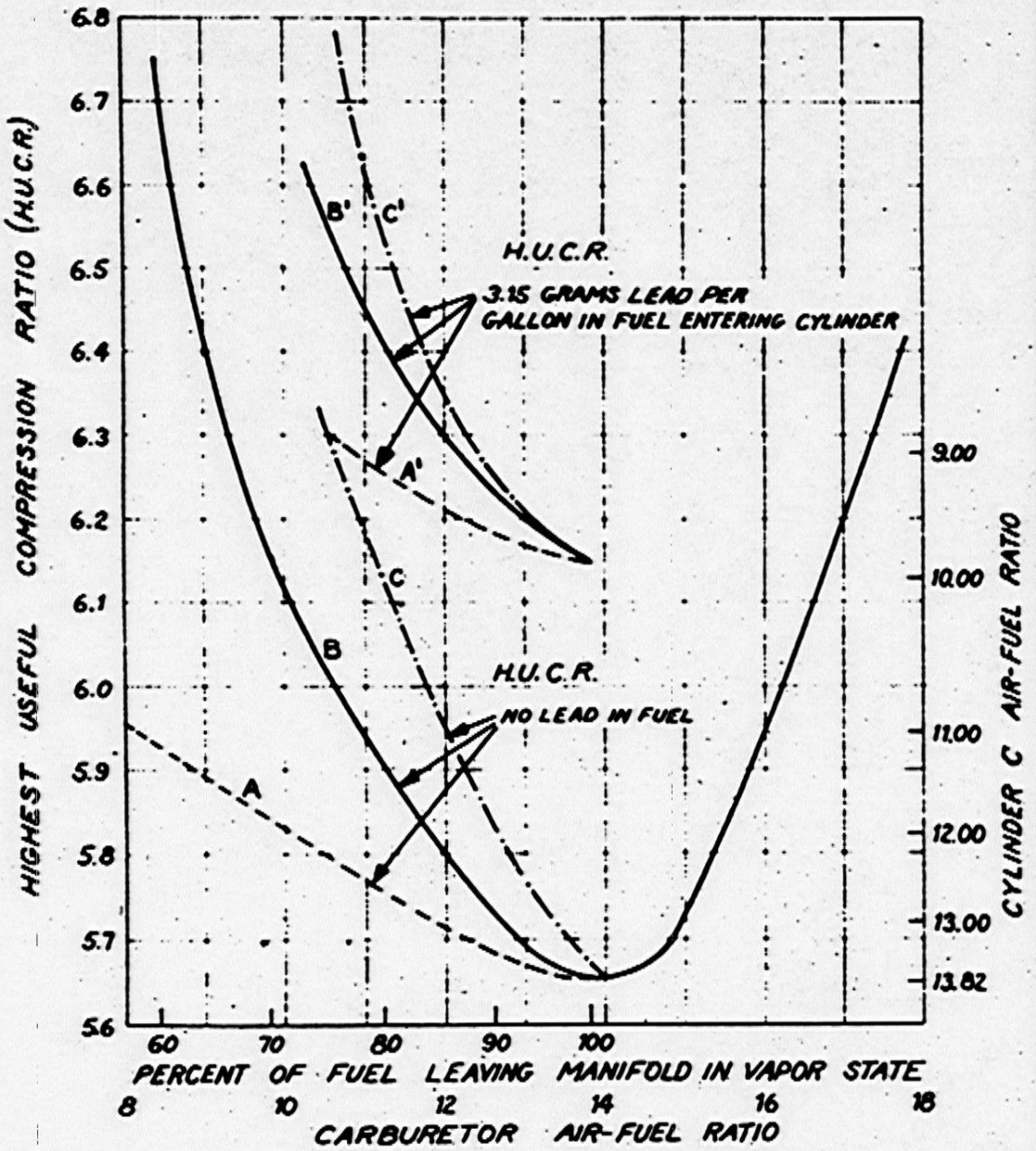
FIG.3.



CYL.	AIR-FUEL RATIO	COMPOSITION OF FUEL ENTERING CYLINDER
A	14.00 TO 1	VAPOR ONLY
B	SAME AS CARBURETOR	SAME AS LEAVING CARBURETOR (VAPOR PLUS NORMAL LIQUID)
C	RICHER THAN CARBURETOR	SAME AS B PLUS NORMAL LIQUID FOR A

CHART (FIG.3)

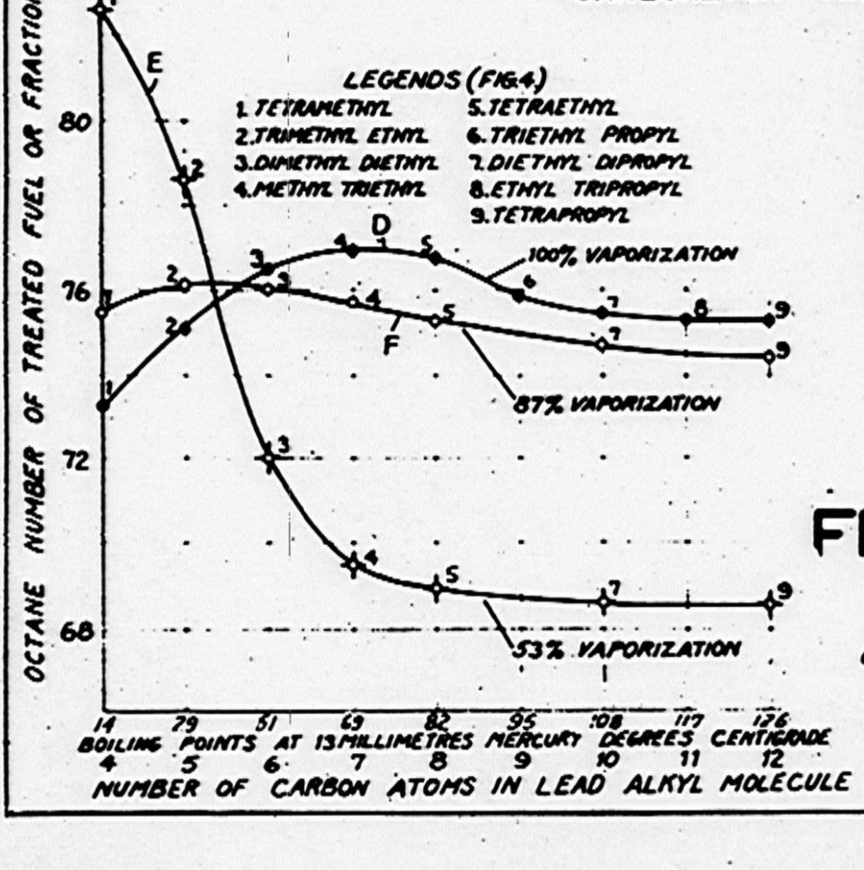


FIG.4.

INVENTOR

Earl Bartholomew

BY

ATTORNEY

188 References cited:

Sullivan	1,938,547	Dec. 5, 1933	44-9.01
Voorhees	1,974,167	Sept. 18, 1934	44-9.01
Alleman	1,949,948	Mar. 6, 1934	44-9.01
Shappirio	2,012,356	Aug. 27, 1936	44-9.01

The claims are all rejected as unpatentable over the references cited which show the use of anti-knock lead alkyls wherein a mixture of different alkyls would be burned simultaneously to give the same effect as applicant's compounds.

At least seven distinct species of anti-knock are present; these are out-lined as follows:

I. Mixture of methyl ethyl lead derivatives (claims 1, 2, 5, 6, 7, 8).

II. Mixture as in I containing also lead tetraethyl (claims 3, 14 and 16).

III. Mixture as in I containing also some lead tetramethyl (claims 4, 9, 10, 15).

IV. Mixture as in I containing also both some lead tetraethyl and some lead tetra methyl (claim 12).

V. Mixture as in I containing also some organic halide (claim 13).

VI. Mixture as in V containing also some lead tetraethyl (claims 14 and 18).

VII. Mixture as in V containing also some lead tetra methyl (claims 14 and 17).

189 The applicant is required to limit the total species of invention as outlined to not more than three according to the provisions of rule 41. The applicant is further required to present an election of one species for further prosecution herein since no generic claim has been allowed order 3254; ex parte Doane 410 O. G. 275.

Claim 19 is rejected as informal.

The claims are further rejected as based upon an inadequate disclosure which fails to accurately describe how the specific compounds claimed are prepared.

A M NASH
Examiner.

* * * * * * * * * *

191 New York, N. Y. June 2, 1937

Commissioner of Patents,
Washington, D. C.

Sir:

This amendment is in response to the Office Action of December 15, 1936.

Cancel claims 13, 14, 17 and 18 constituting groups V, VI and VII, and claim 19.

Remarks:

The basis for the rejection of the claims is not clear from the wording of the rejection. Midgley patent no. 1,573,846 discloses that lead alkyls are antiknocks. A mixture of lead alkyls must also be an antiknock. This follows from the Midgley patent without citation of other references. The patents to Sullivan, Voorhees and Shappirio are directed to methods of making antiknock compounds in hydrocarbon fuels and the mixture resulting from such manufacture. However, these patents do not give any disclosure as to the relative values of antiknock compounds and in this respect they do not add to the above discussion of Midgley's prior patent. The present application 192 claiming certain mixtures of certain lead alkyls requires, as a basis, a statement of relative values which is not found in the references. It appears that the spirit of the rejection is that applicant is claiming merely a mixture of known antiknock compounds but such rejection is untenable in view of the basis laid in the specification and the scope of the claims. If the rejection is repeated the Examiner is asked to apply the references and in doing so will he kindly note in the specification the definition of terms as used in the claims.

The patent to Alleman 1,949,948 is limited to a disclosure of an invention as stated on page 1, lines 21 to 25 inclusive. This patent and the present application follow different bases of thought and they arrive at a different conclusion by a different route. Applicant fails to find in this patent grounds for rejecting the claims.

As to all the references cited, applicant does not find any statement on engine operation and the Examiner's attention is directed to the part of the present application treating engine operation and the part it takes in the present invention.

Acting under the provisions of Rule 41 applicant elects the three species of claims as those of Types II, III and IV as outlined in the rejection. Acting under Order 3254 applicant elects claims of Type II which will be prosecuted if a generic claim is not finally allowed.

Replying to the last paragraph of the rejection it is understood that methods for manufacturing compounds need not be disclosed if they are a part of public knowledge. The following publications give methods of making mixed alkyl compounds of lead:

193 $PbMe_3Et$ — Grüttner and Krause, Berichte der Deutschen Chemischen Gesellschaft (Verlag Chemie, Berlin) 49,1125-33 (1916)
$PbMe_2Et_2$ — “ “ “ “ “ “ “ 49, 1546-50 (1916)
$PbMeEt_3$ — “ “ “ “ “ “ “ 49, 1125-33 (1916)

Methods for making the tetramethyl and tetraethyl compounds of lead are so well known in the literature and including patents that it is believed that the rejection is directed to the mixed alkyl compounds of lead. If desired applicant can also supply citations from the literature relative to making these two compounds.

A more complete description of numerous mixed alkyl lead compounds, together with literature references to their actual preparation, will be found in "The Organic Compounds of Lead" by George Calingaert, Chem. Reviews, Vol. 2, page 43 (1925), published by William and Wilkins, Baltimore.

It is submitted that the application is in condition for allowance.

Respectfully submitted,

EARL BARTHOLOMEW

By JOE W. MORRISON
His Attorney

* * * * * * * * * *

194 In response to amendment filed June 3, 1937.

The claims stand rejected as unpatentable over certain references of record, Sullivan, Voorhees, Alleman and Shappirio. These references show addition to gasoline, or production in situ in the fuel, of mixtures of lead alkyls as anti-knock re-agents which would include lead alkyl compounds of widely varying boiling point range and quantities which of course would be variously distributed to the several cylinders of a motor in operation.

Applicant proposes to use a mixture of low volatile anti-knock substances of varying degrees of volatility to cause more even distribution with the fuel-air mixture to the multiple cylinders of a motor so that each cylinder will receive some lead alkyl throughout the various conditions of low or high motor speeds. Applicant's anti-knock does not completely volatilize, but the use of a mixture of antiknock substances is considered because of its varying degrees of volatility to tend to be more satisfactorily distributed. Such condition and *principle* is met in the references who use lead alkyls in admixture. The references of course do not pick and choose from all of the possible lead alkyls and hence do not specifically state that so much of this lead

alkyl is used and so much of that, but inasmuch as all of the lead alkyls are well known anti-knocks, even as shown by Midgely, and those which applicant's selects are, as he points out, part of the public knowledge, the refer-
195 ences relied on showing use in admixture of the whole gamut of lead alkyls are considered to teach the principle of the invention herein alleged and properly applied to reject the claims even those reciting specific mixtures not directly readable thereon.

The use by these references of a mixture of lead alkyls of widely varying degrees of volatility in various fuels also widely varying in volatility would inherently give the same effect of more even distribution of anti-knock material to the multiple cylinders. This becomes obvious when it is considered that a commercial fuel per se would vary in volatility from about 75° F. to about 425° F., but special fuels might vary more widely. At the same time, the antiknock lead alkyl even the most volatile lead tetra methyl is relatively non-volatile, boiling well above the more volatile fuel components, i. e., about 230° F., so that the applicant's anti-knock is not truly vaporized. Under such conditions any mixture of lead alkyls of different volatilities and gravities would give a similar cylinder distribution pattern to applicant's specific mixture of lead alkyls.

Applicant contends further that certain claims recite a method of engine operation; that the effect of the anti-knock reagent varies therewith, and the references say nothing of engine operation. The claims in effect recite that an engine is operated with a fuel that would knock except that the fuel has been conditioned with a certain anti-knock dope. As a process, the engine is operated in a usual manner except that the fuel is burned with a particular anti-knock dope. There is nothing in these claims to indicate that engine is operated in any unusual manner or that the addition of anti-knock is varied to suit engine operating conditions. If the claims were so limited, division would be properly required since a different divisible invention from the motor fuel would be claimed, separately classified and searched etc. However, the present 196 claims are to the same invention as the motor fuel.

The references add the anti-knock dope to the fuel for the purpose of controlling the combustion thereof in the operation of an ordinary motor and as such are properly applied to these claims as well as those merely reciting the fuel containing the anti-knock dope, or the dope to be added to the fuel.

In Fig. 4 applicant shows that the addition of ordinary anti-knock to fuel of only 53% vaporization has a much lower anti-knock value than fuel of perfect vaporization. But in the graph Fig. I it is indicated that the lower vaporization fuel has the best anti-knock value, and this is supported by Fig. 3 even for a leaded fuel, obviously a direct contradiction of data.

In Fig. 4 the more highly volatile anti-knock radically raises the anti-knock value of the relatively non-vaporizable fuel, but depresses the anti-knock value of vaporizable fuel. But all fuels are incompletely vaporizable under some conditions i. e., winter or starting condition of motor, and all fuels are used in variable conditions of motor operation. Moreover all fuels vary widely in inherent anti-knock characteristics and lead susceptibility and volatility. Hence applicant's claims to a fuel broadly or a motor operated therewith and treated with a dope of variable composition to be used under variable conditions are unduly broad and indefinite, and as held, unpatentable over the prior art.

Summary:

All claims are rejected as unpatentable over Sullivan, Voorhees, Alleman or Shappirio showing motor fuels containing equivalent mixed lead alkyl compounds in view of applicant's admission that his specific alkyls are old.

All claims are rejected as unduly broad and indefinite.

P M NASH
Examiner.

* * * * * * * * * *

204 In response to argument filed Feb. 15, 1938.

Claims 1-12, 15 and 16 are rejected as being indefinite in form and scope as to fail to clearly define the invention disclosed.

The following claim found to be allowable is suggested for the purpose of interference. Applicant should make the same by April 16, 1938 under the provisions of rule 96; failure to do so will be considered a disclaimer of the subject matter involved.

"Liquid fuel for internal combustion engines of the otto cycle multicylinder type comprising a mixture of hydrocarbons of such varying volatility and forming such varying air-fuel ratios as to cause material variation in knocking tendency of the several cylinders in operation and containing a small quantity of a mixture of volatile lead alkyls consisting of tetra ethyl lead, tri eythl methyl lead, di ethyl di methyl lead, ethyl tri methyl lead, and tetra methyl lead, whereby variation in separate cylinder knocking tendency is eliminated and the anti-knock value of the fuel is raised."

P. M. NASH,
Examiner.

* * * * * * * * * *

207 Responsive to argument filed September 2, 1938.

Claims 1-12, 15-16 are rejected upon the disclaimer implied by failure to copy and adopt the claim suggested in the Official action of March 23, 1938, for purposes of interference under the provisions of rule 96.

The claims are again rejected as indefinite as to form and scope as held of record. It is not enough, according to R. S. 4888, that applicant clearly disclose the invention, applicant must distinctly claim it. Often coined expressions to define an artificial class are permitted in claims after laying a proper basis in the disclosure, but the "methyl-ethyl lead derivatives" is not such coined expression, but a term having basis to define a class recognized generally among chemists. Such term for example includes compounds other than those defined in the specification, such as those particularly defined by Alleman, like methyl ethyl diamyl lead. The fact is, therefore, that applicant's claims read upon methyl ethyl lead derivatives other than those disclosed, inoperative for purposes herein, and no reference to the limited definition in the specification is proper and warranted under R. S. 4888, particularly in claims in an application capable of amendment to properly define the invention.

208 Claims 15 and 16 also fail to define the present invention in merely reciting a product unlimited to use in the present invention as a motor fuel anti knock agent. These compositions have no broader utility.

This action is FINAL. Ex parte Hoogendam 499 O. G. 3.

P. M. NASH,
Examiner.

* * * * * * * * * *

209 New York, N. Y. May 12, 1939

Honorable Commissioner of Patents,
Washington, D. C.

Sir:

This is in response to the Office Action of February 7, 1939.

Please amend the above entitled application as follows:

Claim 1, line 6, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 2, line 6, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 3, lines 6 and 7, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

210 Claim 4, lines 6 and 7, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof

—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 5, line 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 6, lines 2 and 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 7, line 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 8, lines 2 and 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 9, line 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl, di-ethyl, and lead tri-ethyl-methyl—.

Claim 10, lines 2 and 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 12, lines 2 and 3, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

211 Claim 15, line 1, cancel "product" and substitute in lieu thereof—fuel anti-knock composition—. Lines 1 and 2, cancel "the three methyl-ethyl lead derivatives" and substitute in lieu thereof—lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

Claim 16, line 1, cancel "product" and substitute in lieu thereof—fuel anti-knock composition—.

REMARKS:

For convenience the claim suggested in the Official Action of March 23, 1938 is quoted herein:

"Liquid fuel for internal combustion engines of the otto cycle multicylinder type comprising a mixture of hydrocarbons of such varying volatility and forming such varying air-fuel ratios as to cause material variation in knocking tendency of the several cylinders in operation and containing a small quantity of a mixture of volatile lead alkyls consisting of tetra ethyl lead, tri ethyl methyl lead, di ethyl di methyl lead, ethyl tri methyl lead, and tetra methyl lead, whereby variation in separate cylinder knocking tendency is eliminated and the anti-knock value of the fuel is raised."

Applicant raises two points in answer to the rejection.

First: Applicant cannot find and the Examiner has not pointed out any subject matter in the present application which will support this claim or which states that applicant is trying to achieve the result claimed. Applicant specifically denies that this claim names a direction or a result which the present application endeavors to attain.

* * * * * * * * * *

216 Responsive to amendment filed May 15, 1939.

Applicant contends that the final rejection herein is premature on the ground that objection to the term "product" was not raised earlier in claims 15 and 16 and that applicant was given no opportunity to be heard upon the disclaimer. In view thereof the final character of the rejection is withdrawn.

The claims appear to be in proper form as amended and this ground of rejection is withdrawn.

The rejection of all claims herein upon the disclaimer under rule 96 is repeated. Applicant contends that neither he nor any one else could make a claim of the terminology of the claim suggested under rule 96.

The specific terms objected to is the final functional "whereby" clause defining an effect which applicant contends is impossible and inoperative. Since the proposed claim otherwise does define the substance of applicants motor fuel product and would otherwise function in the motor

the same way, in whatever terms this function may be described, it is considered that applicant was not warranted in disclaiming the same because the present claims define no patentable substance thereover; that applicant should have made this claim, otherwise properly defining his motor fuel product and anti knock dope, and objected to what-
217 ever terms therein he conceived to be objectionable interpartes during the motion period as provided in Rule 122 and suggested a claim better suited, if he desired, as provided in rule 109.

Since applicant did not do this it is considered that the proposed claim is proper prior art herein for reasons pointed out.

This rejection is now made FINAL the period of response to which runs from the date of this letter.

P. M. NASH,
Examiner.

* * * * * * * * * *

220 *Examiner's Statement*

This is an appeal from the final rejection of claims 1-12, 15 and 16 upon the ground that the subject matter thereof is disclaimed under rule 96 for failure to make a count suggested herein for interference.

The claims on appeal are:

1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression of the motor fuel and burning the mixture together within a primary anti-knock agent comprising a mixture of lead trimethyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl—.

2. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compres-

sion pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl in which antiknock mixture the lead dimethyl diethyl predominates.

3. The method of operating an internal combustion engine comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead dimethyl di-ethyl, and lead tri-ethyl-methyl and lead tetraethyl the concentration of the lead being greatest in the lead triethyl methyl and lead tetraethyl.

4. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl and lead tetramethyl in a concentration not exceeding fifteen percent.

221 5. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

6. A product comprising a motor fuel and a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

7. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, in which mixture the concentration of lead dimethyl diethyl predominates.

8. A product comprising a motor fuel and a primary antiknock agent consisting of lead tri-methyl-ethyl, lead dimethyl di-ethyl, and lead tri-ethyl-methyl, in which the concentration of lead dimethyl diethyl predominates.

9. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, and lead tetramethyl in a concentration not exceeding fifteen percent.

10. A product comprising a motor fuel and a primary antiknock consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, and lead tetra-methyl in a concentration not exceeding fifteen percent.

11. A product comprising a motor fuel and an antiknock agent comprising lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl and in which the greatest lead concentration is in the group consisting of lead triethyl methyl and lead tetraethyl.

12. A product comprising a mixture of a motor fuel, a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetramethyl and lead tetraethyl present in relatively small concentrations.

15. A fuel and antiknock composition comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl and lead tetramethyl in a concentration not exceeding fifteen percent.

16. A fuel anti-knock composition comprising a mixture of lead tri-methyl ethyl, lead dimethyl diethyl, lead triethyl methyl and lead tetraethyl with the greatest lead concentration in the lead triethyl methyl and lead tetraethyl.

The proposed claim under rule 96 is copied:

222 "Liquid fuel for internal combustion engines of the otto cycle multicylinder type comprising a mixture of hydrocarbons of such varying volatility and forming such varying air-fuel ratios as to cause material variation in knocking tendency of the several cylinders in operation

and containing a small quantity of a mixture of volatile lead alkyls consisting of tetra ethyl lead, tri ethyl methyl lead, di ethyl di methyl lead, ethyl tri methyl lead, and tetra methyl lead, whereby variation in separate cylinder knocking tendency is eliminated and the anti-knock value of the fuel is raised."

Other references of record:

Sullivan	1,938,547	Dec. 5, 1933
Voorhees	1,974,167	Sept. 18, 1934
Shappiro	2, -12,356	Aug. 27, 1936

The invention disclosed is concerned with improvement of motor fuels such as gasoline with respect to its tendency to knock in multicylinder internal combustion engines by addition of lead alkyls as anti-knock dope. Applicant proposes use of certain mixtures of lead alkyls as anti-knock.

The prior art listed shows the use of lead alkyls in admixture and applicant admits that it lies within the skill of the art to use ordinary mixtures of lead alkyls as anti-knock dope.

Applicant's mixture is one of adjusted volatility and anti-knock effect thru use of mixtures of lead methyl and ethyl derivatives such as lead tetraethyl, lead tetramethyl and intermediate alkyls, lead tri-ethyl-methyl, di-ethyl di-methyl and tri-methyl-ethyl.

In ordinary carburetion of an air-fuel mixture comprising a mixture of air, fuel vapor and unvaporized suspension therein of liquid fuel particles, inertia effects upon the suspended particles of variable distances the mixture travels to the several cylinders, variable speeds of operation
223 as rapid acceleration, starting, stopping as well as
temperature variation, will give variation in the fuel-air charge actually received by individual cylinders. This variation results in different knocking tendencies of the several cylinders.

By using a mixture of lead alkyls of variable volatility and anti-knock value to be added to the fuel, applicant contends that this condition is improved due at least in part to better distribution of anti-knock under such variable conditions.

As the examiner understood the invention, a claim as copied above was formulated to express the same and proposed under rule 96 to applicant and other parties. The claim was adopted by other parties and this claim has become the basis of an interference contest, but applicant refused to make this claim from which action the present rejection of all claims upon the rule 96 disclaimer stems.

Applicant argues that the proposed claim is inoperative and he has no basis therefor, at least in the functional clause,

> ''whereby variation in separate cylinder knocking tendency is eliminated—.''

Therefore he refused to make the claim and contends that his present claims substantially amended should be allowed.

It is the Examiner's position that the suggested count is not inoperative, does express the invention in all *material aspects,* and should have been made at least for purposes of interference and any objection thereto raised inter-partes or the burden of formulating better claims accepted as provided by the interference rules, hence all claims herein are deemed properly rejected on the disclaimer.

In the Examiner's view a claim suggested for the purpose of interference is at least notice to an applicant that interfering applications exist in the Patent Office, claiming common subject matter and the proposed claims formulated by the Examiner are not necessarily the exact claims of any of the prospective parties and are subject to inter-
224 partes modification according to the rules.

Basis is found for applicant's present claims in the applications of other parties for which applicant may not be the first inventor and the claims therefore cannot be allowed to him without a contest of priority thereon, and under rule 96, the refusal of interference by disclaimer is deemed to be a disclaimer of all common subject matter and in such case the validity of the claim actually suggested is not an issue.

Considering however the merits of the contention that the claim suggested is invalid, it is noted the claim is drawn to a motor fuel composition stated to have certain properties. By extreme interpretation applicant contends that his composition does not have such property. However, applicant discloses the same composition as defined in the proposed claim and uses it in the same way. The properties thereof are inherent and will be the same in all cases however described. Certainly a motor fuel composition as disclosed by applicant cannot be patentable over a composition suggested by the Examiner because applicant argues that his composition will not "eliminate the separate cylinder knocking tendency".

Thus it is noted that irrespective of the final functional "whereby" clause, the claim does define the motor fuel composition disclosed by applicant and therefore defines all that is material in applicant's invention.

But considering applicant's argument more specifically that the claim is inoperative because it is not possible to "eliminate the separate cylinder knocking tendency", it can only be pointed out that the dope comprising the mixed lead alkyls of different volatilities is an adjustment in this direction. Applicant only uses the mixed lead alkyls because they have different volatilities and the lighter ones under certain conditions volatilize better and tend to overcome the knocking tendency in cylinders of increased air-fuel ratio (abnormally lean mixtures). The more volatile lead alkyls have decreased intrinsic anti-knock value as compared to other lead alkyls. They are therefore used only admixed with other less volatile better anti-knocks. In short, the more volatile anti-knocks are used with due appreciation of their low anti-knock character in admixture with better anti-knock. But applicant admits as shown by the art that the use of mixtures of lead alkyls is old, admits that each of his specific lead alkyls is old, and emphasizes throughout that no lead alkyl alone or in admixture is as good in *intrinsic* anti-knock value as lead tetraethyl per se.

How then can anything involve invention herein except the specific combination of lead alkyls in admixture to solve a specific problem?

Applicant's specification is a scientific treatise on the separate cylinder knocking tendency due to poor fuel distribution.

This is the problem applicant solves and he does this by use of a mixture of lead alkyls superior to solve this problem, because the separate cylinders get the anti-knock dope when they need it, not withstanding none of the dope would be as good as lead tetraethyl per se in a perfect fuel distribution system.

Thus the mixture of alkyls is most important to eliminate the separate cylinder knocking tendency, which varies with different operating conditions, while incidently raising the anti-knock value of the fuel overall. *This is what the suggested claim defines. This is also what applicant says is inoperative*—he cannot eliminate the separate cylinder knocking tendency.

226 Applicant interprets the "whereby" clause to mean that each cylinder charge must have exactly the same anti-knock value, but this is beyond any reasonable interpretation of the claim. Two cylinders of different fuel charges of different anti-knock hydrocarbon types, of different air-fuel ratios and of different lead alkyls anti-knock dope content could rarely have the same anti-knock value. For purposes of the claim it is sufficient that neither cylinder knocks under the ordinary range of operating conditions of the motor. This is what applicant says in effect (page 18, 2nd paragraph).

If a motor fuel were doped with a mixture of lead alkyls of the type defined herein for the purpose of rendering it anti-knock in all cylinders over the whole range of usual operating conditions of a multicylinder motor, the terms of the suggested claim would be met. Hence such strained interpretation that each cylinder must attain the absurd condition that it has exactly the same tendency to resist knocking as all the others of a multicylinder engine is not justification for the disclaimer herein.

It is summarized therefore—

(1) that applicant has basis for all limitations in a motor fuel composition claim suggested under rule 96 for interference and disclaimed;

(2) that the present claims do not patentably distinguish the disclaimed matter;

(3) the present claims could not be allowed to applicant at least for the reason that he may not be the first inventor thereof and has already refused a contest of interference upon not patentably distinguishable subject matter by disclaimer.

227 It is considered to be properly held that all claims herein are rejectable upon this disclaimer.

Respectfully,

P. M. NASH,
Examiner.

* * * * * * * * * *

256 Endorsed: U. S. Patent Office Board of Appeals Oct 15 1940 Mailed

In the United States Patent Office

Before the Board of Appeals

Ex parte Earl Bartholomew

Application for Patent filed September 4, 1936, Serial No. 99,497. Method and Means for Improving Engine Operation.

Mr. Joe W. Morrison for applicant.

This is an appeal from the final rejection of all claims remaining in the application, namely, 1 to 12, 15 and 16.

Claims 1, 3, 4, 5, 9 and 11 are illustrative.

1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead triethyl-methyl.

3. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead triethyl-methyl; and lead tetraethyl the concentration of the lead being greatest in the lead triethyl methyl and lead tetraethyl.

257 4. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-thyl, and lead tri-ethyl-methyl; and lead tertamethyl in a concentration not exceeding fifteen percent.

5. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

9. A product comprising a motor fuel and an antiknock material having as its primary antiknock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, and lead tetramethyl in a concentration not exceeding fifteen percent.

11. A product comprising a motor fuel and an antiknock agent comprising lead tri-methyl ethyl, lead di-methyl di-

ethyl, lead triethyl methyl and lead tetraethyl and in which the greatest lead concentration is in the group consisting of lead triethyl methyl and lead tetraethyl.

The patents cited by the examiner are as follows:

Sullivan et al	1,938,547	Dec. 5, 1933
Voorhees	1,974,167	Sep. 18, 1934
Shappirio	2,012,356	Aug. 27, 1935

The appealed claims relate to the matter of reducing knock in internal combustion engines employing gasoline. As may be noted from the quoted claims, the subject matter is set forth first as a method comprising a mixture with a fuel charge of certain lead alkyl compounds and in the second place as a fuel containing such lead alkyls. Applicant employs the five possible lead compounds of methyl and ethyl including mixtures. He prefers employment of the three mixed alkyls but may include the tetra compound of both methyl and ethyl.

The claims have been rejected as not patentably different from a claim that was suggested under the provisions of Rule 96 for purpose of interference and which applicant refused to make and in connection with the citations. 258 Certain citations of the prior art are shown as illustrative of the status of this subject matter in this antiknock art.

The suggseted claim which was embodied in the examiner's letter of March 23, 1938 is as follows:

Liquid fuel for internal combustion engines of the otto cycle multicylinder type comprising a mixture of hydrocarbons of such varying volatility and forming such varying air-fuel ratios as to cause material variation in knocking tendency of the several cylinders in operation and containing a small quantity of a mixture of volatile lead alkyls consisting of tetra ethyl lead, tri ethyl methyl lead, di ethyl di methyl lead, ethyl tri methyl lead, and tetra methyl lead, whereby variation in separate cylinder knocking tendency is eliminated and the anti-knock value of the fuel is raised.

The quoted claim includes all five of the possible combinations of lead with methyl and ethyl and their mixed forms.

It is contended on behalf of applicant that the proposed claim does properly represent his invention and further, that it cannot be read upon applicant's disclosure and statement of invention, that applicant regards the features of the appealed claims to be inventive and patentably different from the proposed claim. We regard this as the question presented for determination here.

Much discussion is presented by applicant in respect to avoiding or eliminating variation in separate cylinder knocking tendency rather than merely broadly reducing knock. Applicant illustrates by drawing and disclosure how this is said to be attained involving largely a selection and adjustment as between the several lead alkyl members.

259 In treating the situation we shall assume that the suggested claim together with the three citations and the patent to Midgley 1,573,846 discussed in applicant's brief as representing, in effect, the state of the art, are available as references. It seems clear upon inspection of the situation generally that the claim suggested for interference by applicant is so close to and doubtfully different from the disclosure of applicant's case that applicant entailed the consequence of Rule 96 in not making the same.

We have carefully considered the matters of merit of the disclosure in connection with such above noted prior art or grounds of rejection and conclude that we can see no sufficient line of patentable difference to warrant allowance of the appealed claims thereover. Taking the total of the available prior art including the suggested claim, it is our conclusion that the appealed claims represent only matter of routine test and choice of one or more or combinations of the several available lead alkyls of the suggested claim. We believe the patents are suggestive of trying various selected mixtures of lead alkyls though admittedly not of exactly the limitations of the appeal claims. However, we regard the difference as not involving invention.

We find no error in the examiner's conclusions as based mainly upon the suggested claim in one instance or as upon this claim in view of the collateral showing of the state of art.

The decision of the examiner is affirmed.

BOARD OF APPEALS

EUGENE LANDERS,
Examiner-in-Chief
W. L. REDROW
Examiner-in-Chief
B. B. PIERCE
Examiner-in-Chief

October 15, 1940

* * * * * * * * * *

268 *On Request for Clarification and Reconsideration*

In response to petition for clarification and reconsideration filed November 7, 1940 of our decision dated October 15, 1940.

It was not intended to raise any new ground of rejection in our decision. The state of the art was referred to merely as background of the situation, the same as, but to no greater extent than was done by the examiner in the paragraph, middle of page 3 of his answer.

Reopening of the prosecution is not approved.

The features re-emphasized in the additional remarks were given full consideration in reaching the conclusions stated and have been reconsidered without finding reason for changing our decision.

269 No change is necessary in the sentence of our decision discussed on page 3 of the remarks. The clause by applicant modifies the word "interference" and not the word "suggested" as assumed by counsel.

For the reasons noted in our decision and herein the decision of the examiner is affirmed.

BOARD OF APPEALS

EUGENE LANDERS,
Examiner-in-Chief
W. L. REDROW
Examiner-in-Chief
B. B. PIERCE
Examiner-in-Chief

Mr. Joe W. Morrison,
c/o Ethyl Gasoline Corp.,
Chrysler Bldg.,
New York, N. Y.

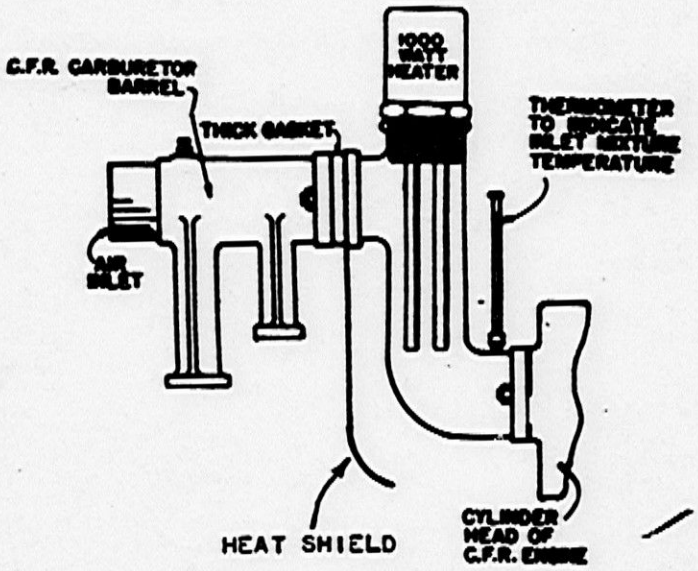
November 14, 1940.

* * * * * * * * * *

Ti's Exhibit 2.

FILED
NOV 13 1942
CHARLES E. STEWART, Clerk

FILE REF. NO. 92.3030 6020


1000 WATT HEATER
C.F.R. CARBURETOR BARREL
THICK GASKET
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
AIR INLET
HEAT SHIELD
CYLINDER HEAD OF C.F.R. ENGINE

Coop Fuel R. Comtee Engine

INDUCTION SYSTEM FOR THE C.F.R. ENGINE, AS USED IN MAKING A.S.T.M. MOTOR METHOD KNOCK RATINGS

ETHYL GASOLINE CORPORATION ENGINEERING LABORATORIES DETROIT, MICHIGAN	DRAWN BY B.B.	DRAWING NO. S'41-5002

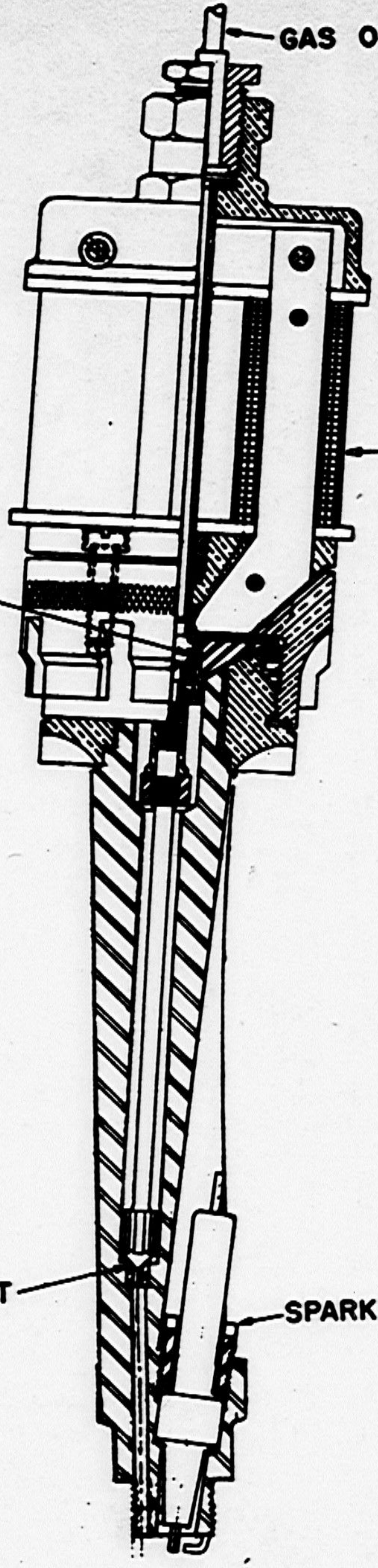


FIG.1 VALVE FOR SAMPLING MIXTURE IN THE CYLINDERS OF A MULTICYLINDER ENGINE.

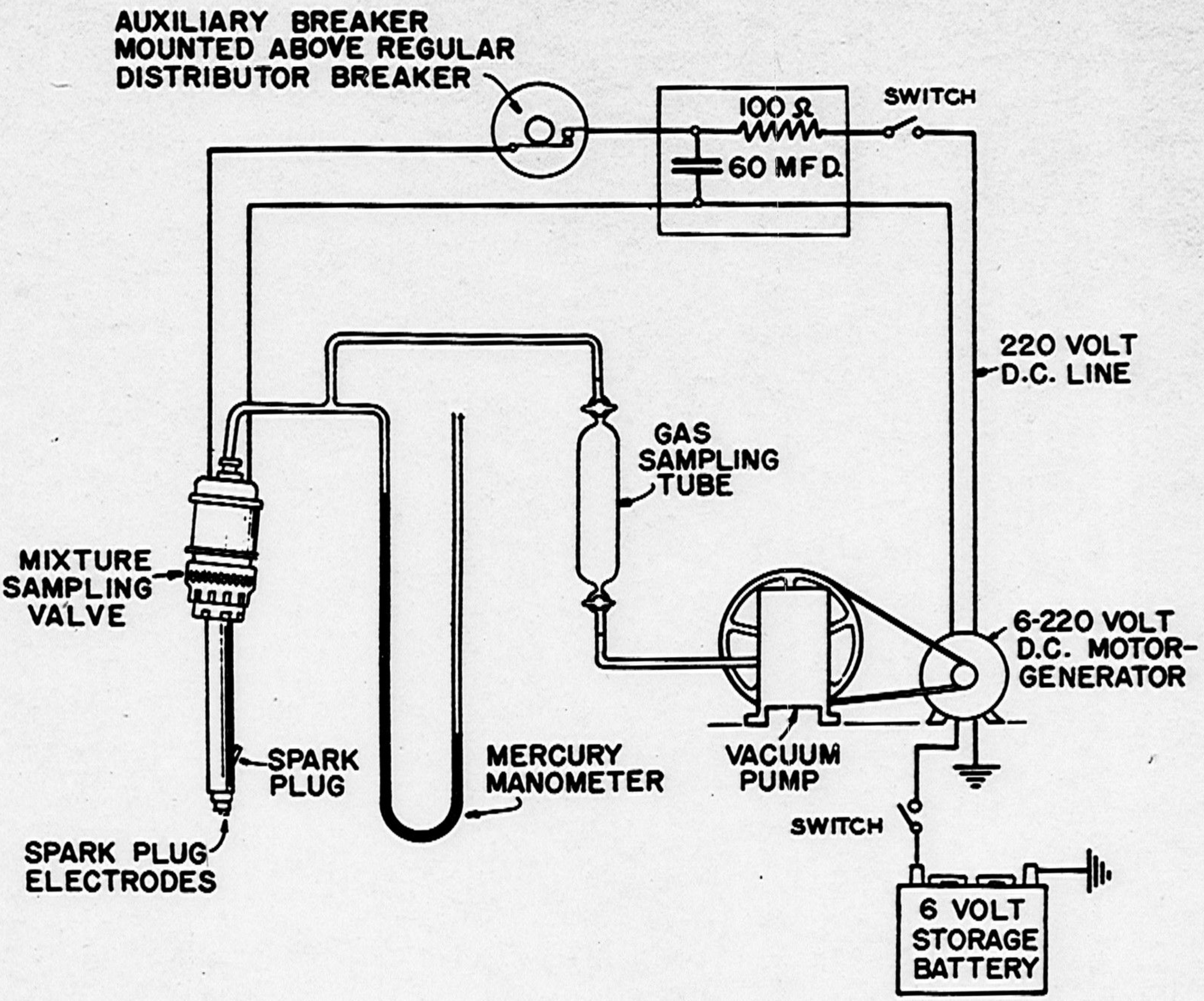


FIG. 2 DIAGRAMMATIC SKETCH OF MIXTURE SAMPLING VALVE AND ASSOCIATED EQUIPMENT.

Exhibit 5

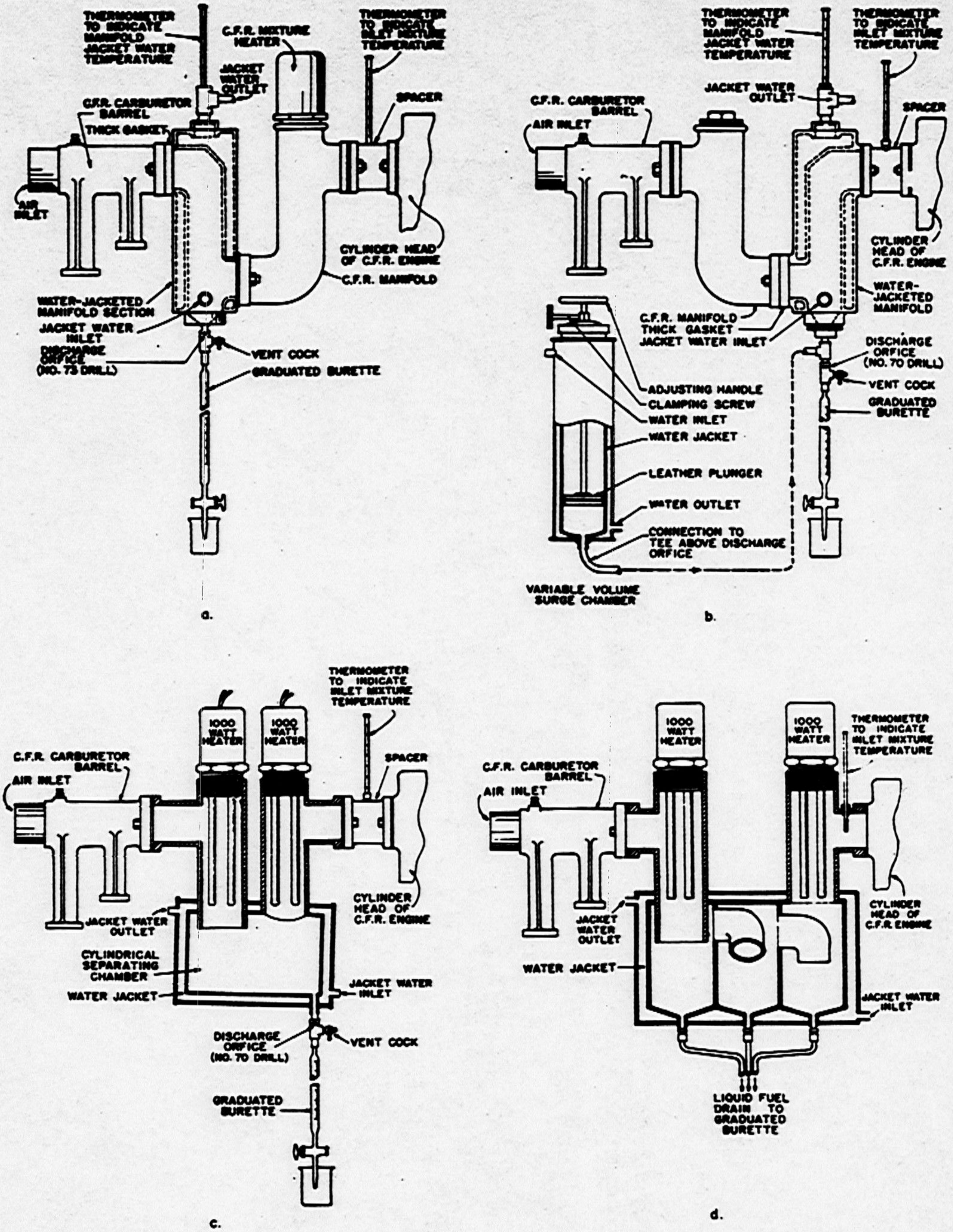

THERMOMETER TO INDICATE MANIFOLD JACKET WATER TEMPERATURE
C.F.R. MIXTURE HEATER
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
JACKET WATER OUTLET
C.F.R. CARBURETOR BARREL
THICK GASKET
SPACER
AIR INLET
CYLINDER HEAD OF C.F.R. ENGINE
C.F.R. MANIFOLD
WATER-JACKETED MANIFOLD SECTION
JACKET WATER INLET
DISCHARGE ORFICE (NO. 73 DRILL)
VENT COCK
GRADUATED BURETTE
a.
THERMOMETER TO INDICATE MANIFOLD JACKET WATER TEMPERATURE
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
JACKET WATER OUTLET
C.F.R. CARBURETOR BARREL
AIR INLET
SPACER
CYLINDER HEAD OF C.F.R. ENGINE
WATER-JACKETED MANIFOLD
C.F.R. MANIFOLD
THICK GASKET
JACKET WATER INLET
DISCHARGE ORFICE (NO. 70 DRILL)
VENT COCK
GRADUATED BURETTE
ADJUSTING HANDLE
CLAMPING SCREW
WATER INLET
WATER JACKET
LEATHER PLUNGER
WATER OUTLET
CONNECTION TO TEE ABOVE DISCHARGE ORFICE
VARIABLE VOLUME SURGE CHAMBER
b.
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
1000 WATT HEATER
1000 WATT HEATER
C.F.R. CARBURETOR BARREL
AIR INLET
SPACER
CYLINDER HEAD OF C.F.R. ENGINE
JACKET WATER OUTLET
CYLINDRICAL SEPARATING CHAMBER
WATER JACKET
JACKET WATER INLET
DISCHARGE ORFICE (NO. 70 DRILL)
VENT COCK
GRADUATED BURETTE
c.
1000 WATT HEATER
1000 WATT HEATER
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
C.F.R. CARBURETOR BARREL
AIR INLET
CYLINDER HEAD OF C.F.R. ENGINE
JACKET WATER OUTLET
WATER JACKET
JACKET WATER INLET
LIQUID FUEL DRAIN TO GRADUATED BURETTE
d.

FIG.3 DIAGRAMMATIC SKETCHES OF INTAKE MANIFOLDS FOR THE C.F.R. ENGINE WHICH SIMULATE THE EFFECTS OF IMPERFECT MIXTURE DISTRIBUTION IN THE MULTICYLINDER ENGINE.

FILED
NOV 13 1942
CHARLES E. STEWART, Clerk

Carburetion, Manifolding and Fuel Antiknock Value

By Earl Bartholomew, Harold Chalk, and Benjamin Brewster
Ethyl Gasoline Corp.

INCREASED performance at high speed, which is an important characteristic of cars built during the last few years, has required the use of intake manifolds of considerably greater cross-sectional area which, together with certain other changes in engine design, cause unequal distribution of the less volatile portions of the fuel not vaporized in the manifold. Consequently, the fuel which enters the cylinders varies in composition. If other conditions are equal, the relative knocking tendency of the cylinders is determined by the distribution of antiknock value through the fractions of the fuel and by the air-fuel ratios of the mixtures in the cylinders. Ordinarily the leanest cylinder knocks the most.

Fuels rated in the laboratory by conventional methods as having equal antiknock value may differ considerably in road performance because of differences in volatility and distribution of antiknock value through their fractions. The problem is complicated further by differences in the volatility of the commonly used secondary reference fuels.

Recently developed instrumentation permits the determination of the knocking tendency, air-fuel ratio, and spark advance for the individual cylinders of an automobile engine on the road. A modified fuel system and intake manifold for the C.F.R. engine make possible the duplication and measurement in the laboratory of the effects caused by imperfect fuel distribution within the car engine.

DURING the past six years the development of laboratory testing methods to give knock ratings that correlate with the ratings obtained in cars on the road has been a major problem of the oil refiner and car manufacturer. After the cooperative road-testing program at Uniontown, Pa., in 1932 a laboratory procedure was developed which gave ratings agreeing remarkably well with the road ratings. The ratings obtained on the road in the second Uniontown program in 1934, did not agree quite so well with ratings obtained by the Motor Method.

Since that time road ratings have tended to deviate more and more from the consistent pattern which characterized the earlier ratings. This deviation applies to average road ratings and particularly to ratings in individual cars. Fuels which have approximately the same Motor and Research Method ratings may have road ratings as much as 8 octane numbers higher or lower than their laboratory ratings. Fuels which have a fairly large difference between their Research and Motor Method ratings may, on the road, show no appreciation over their Motor Method ratings, and others having the same difference between their Research and Motor Method ratings in the laboratory may, under the same conditions of car operation, have road ratings considerably higher than their Research-Method ratings.

As an example of such performance we may consider the ratings of two specially blended fuels whose road and laboratory ratings are tabulated below:

Table 1 – Road and Laboratory Knock Ratings of Two Specially Blended Fuels

Fuel	Research-Method Rating	Motor-Method Rating	Average of Winter Road Ratings in Eleven Cars	Winter Road Rating in Car No. 155
S-6	72.9	75.6	71.4	67.6
S-11	71.7	73.8	74.8	80.0

The average road rating of fuel S-6 is substantially *lower* than either its Motor- or Research-Method rating, and the average road rating of fuel S-11 is considerably *higher* than either its Motor- or Research-Method rating. Although the Research- and Motor-Method ratings of the two fuels differ by 1.2 and 1.8 octane numbers respectively in one direction, their ratings in Car No. 155 differ by 12.4 octane numbers in the opposite direction.

Car No. 155 is highly satisfactory for knock-testing purposes, being relatively free of noises other than that due to knock and having more than average sensitivity to changes in fuel octane number. The influence of certain engine variables such as speed, spark advance, and temperature on the relative antiknock value of different types of fuels is recognized generally, and may be demonstrated readily in the laboratory on single-cylinder engines such as the A.S.T.M.-C.F.R. engine, but no variations of the Motor and Research Methods which have been tried, involving only changes in speed, temperature, and spark advance, have served to bring the laboratory ratings of these fuels into agreement with the road ratings.

[This paper was presented at the Annual Meeting of the Society, Detroit, Mich., Jan. 14, 1938.]

Changes in Engine Design and Fuels

A study of such data obtained in numerous laboratories during the past three years led to the suspicion that some factor or combination of factors as yet neglected in the standard laboratory knock-testing procedure, in some cases, may have roughly as great an influence on road ratings as have engine speed and temperature. Since the serious disagreement of road ratings and laboratory ratings made by the generally established procedures, or simple variations thereof, is of relatively recent origin, the changes in engine design which may account for such deviations are of immediate interest. The following changes seem to warrant attention:

(1) Increased cross-sectional area of intake manifolds in conjunction with the general adoption of down-draft carburetors having larger tube diameters, resulting in considerably lower intake-manifold velocities in the low-speed range of the engine, to which knocking now is confined generally.

(2) The automatic choke.

(3) Decreased mixture heating by the exhaust gases and the use of automatic heat-control valves whose operation is controlled largely by the temperature of the air surrounding the bi-metallic element.

(4) The elimination of thermostatically controlled radiator shutters.

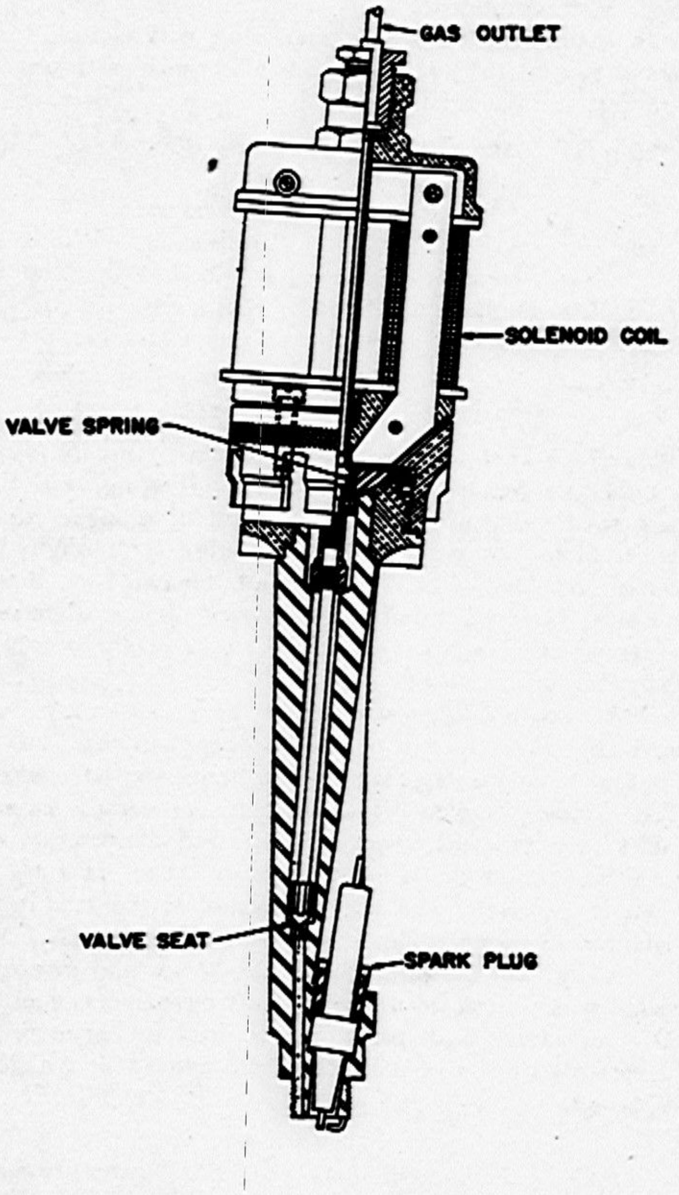


FIG. 1 VALVE FOR SAMPLING MIXTURE IN THE CYLINDERS OF A MULTICYLINDER ENGINE.

In addition to these engine-design factors there is the possible effect of recent developments in gasoline production such as polymerization, reforming, and new cracking methods. It is the purpose of this paper to review some of the recent work in the laboratories of the Ethyl Gasoline Corp. which has had for its purpose:

(1) The explanation for at least some of the previously described unusual performance of fuels in cars on the road.

(2) The development of improved instrumentation for the analysis of events in the intake manifold and cylinder.

(3) The formulation of principles to govern the selection of fuels to meet best any given set of manifold and carburetion conditions.

The paper is designed to present fundamental principles that apply to a complex problem, hence, in the interest of simplification, the discussion is confined in so far as possible to typical conditions to be met.

Campbell, Lovell and Boyd[1] pointed to the probability that mixture ratio and fuel volatility affect the relative antiknock value of fuels and cited tests in cars on the road which appeared to confirm the hypothesis. The work in the laboratories of the Ethyl Gasoline Corp. indicates that fuel volatility, variation of fuel antiknock value with distillation temperature, carburetor air-fuel ratio, design of the intake manifold and associated parts, variation of other conditions which affect fuel distribution within the engine, and the reference fuels used, all have an important bearing on the relative knock ratings of fuels.

Fuel Distribution Within the Engine

Fuel of substantially uniform composition is distributed to all the cylinders of a multicylinder engine in approximately equal proportions with the admixed air if the available heat, time of contact of air with fuel, and the agitation of the mixture are sufficient to vaporize the fuel completely and mix it homogeneously with the air, or if the manifold velocity is high enough to carry the liquid fuel in the air stream by entrainment. High volumetric efficiency of automobile engines has been obtained only with reasonably low intake-manifold velocity. The high brake mean effective pressures at high engine speed in recently built cars require the use of intake manifolds of large cross-sectional area and a minimum of mixture heating. Both of these design features are diametrically opposed to good fuel distribution at low car speeds where knocking has become most prevalent. It is impossible to combine in a single conventional unsupercharged manifold the most satisfactory characteristics for both high- and low-speed operation.

The determination of the air-fuel ratios of the mixtures entering the individual cylinders of a multicylinder engine on the road has always presented a problem. Samples of exhaust gases withdrawn from the exhaust manifold as close as possible to the exhaust valves are invariably contaminated by products of combustion from other cylinders. The substitution for the conventional manifold of a separate exhaust stack for each cylinder destroys the standard hot-spot arrangement and considerably increases the temperature under the hood. The most desirable approach is the sampling of the mixture within each cylinder. This procedure has been made possible by the combination spark-plug and gas sampling valve shown in Fig. 1. The small solenoid valve between the cylinder and the gas sampling tube has only a small movement and remains open for a very short period. Its operation is controlled by an auxiliary breaker mechanism mounted above the conventional distributor and driven by the distributor shaft. At the instant the points of the auxiliary breaker close, a condenser in the circuit discharges a relatively heavy current through the solenoid coil and causes the valve to open momentarily.

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[1] See S.A.E. Transactions, April, 1937, pp. 144-148: "Relative Knocking Characteristics of Motor Fuels in Service," by John M. Campbell, Wheeler G. Lovell, and T. A. Boyd.

At all other times the valve is held in the closed position by the pressure of a coil spring.

The complete apparatus is shown diagrammatically in Fig. 2. When a sample is to be obtained, the vacuum pump, which is driven by a motor which draws its current from a 6-220 volt motor-generator set, evacuates the gas sampling tube to an absolute pressure of about 5 mm. mercury. The car is accelerated under the conditions prevailing in the road knock test and the valve is allowed to operate only within the 5 m.p.h. speed range of maximum knock intensity. From 6 to 8 repetitions of the acceleration are required to fill the gas sampling tube to atmospheric pressure. The procedure is then repeated for the other cylinders.

Table 2 – Air-Fuel Ratios of the Mixtures in the Cylinders of an Automobile Engine at 10 to 15 M.P.H.
Atmospheric Temperature, 56 Deg. Fahr.,
Fuel of Winter Volatility

Cylinder No.							
1	2	3	4	5	6	7	8
14.59	16.01	13.29	14.13	12.63	11.56	15.27	12.37

Average air-fuel ratio, all cylinders	13.6
Average for cylinders 1, 2, 7 and 8	14.4
Average for cylinders 3, 4, 5 and 6	12.8

Table 2 shows the air-fuel ratios for the individual cylinders of a certain car during acceleration at 10 to 15 m.p.h. at an atmospheric temperature of 56 deg. fahr., as determined by the apparatus shown in Fig. 2. The mean air-fuel ratio computed from these data is shown, also the computed means for cylinder groups 1, 2, 7 and 8 and 3, 4, 5 and 6, each of which is fed by a single barrel of a dual down-draft carburetor.

For the same conditions of operation, the mean air-fuel ratio is normally richer than that shown in Table 2. There is, however, always a tendency for a portion of the heavy ends of the acceleration charge to lag behind the air-vapor mixture for a short period after the sudden opening of the throttle, with the result that the mean cylinder air-fuel ratio is momentarily leaner than after the lagging liquid fuel starts to flow into the cylinders. This factor may, in part, account also for the difference between the mean air-fuel ratios for the two groups of cylinders, inasmuch as the manifold for cylinders 1, 2, 7 and 8 is longer than that for the remaining cylinders and has more surface to be wet by the acceleration charge. It should be noted that the rear cylinder of each group receives the richest mixture, presumably because of the forward motion of the car which tends to cause liquid fuel to accumulate in the rear of the manifolds.

The air-fuel ratio of the mixtures in cylinders 1, 2, 7 and 8 varies from 12.37 to 16.01. Often the variation is greater. The mechanism by which fuel and air are delivered to the cylinders in such a wide range of proportions appears to be as follows:

Air and fuel are metered by the carburetor and enter the manifold. The magnitude of the fraction of the fuel in the vapor state as the mixture leaves the manifold is a function of the heat in the fuel and air supplied to the carburetor, the heat added by the manifold, the volatility and latent heat of the fuel, and the efficiency of mixing in the manifold. Although the available heat may be adequate to ensure complete vaporization under equilibrium conditions, the time of contact and degree of mixing in the manifold may not be sufficient to bring about this condition.

The air and the lighter fractions of the fuel, vaporized in the carburetor and manifold, are distributed substantially equally to the cylinders. A part of the unvaporized, less volatile portion is distributed fairly uniformly by entrainment with the air, but the quantity of heavy ends so distributed becomes less as engine speed decreases and appears to be small at low engine speed.

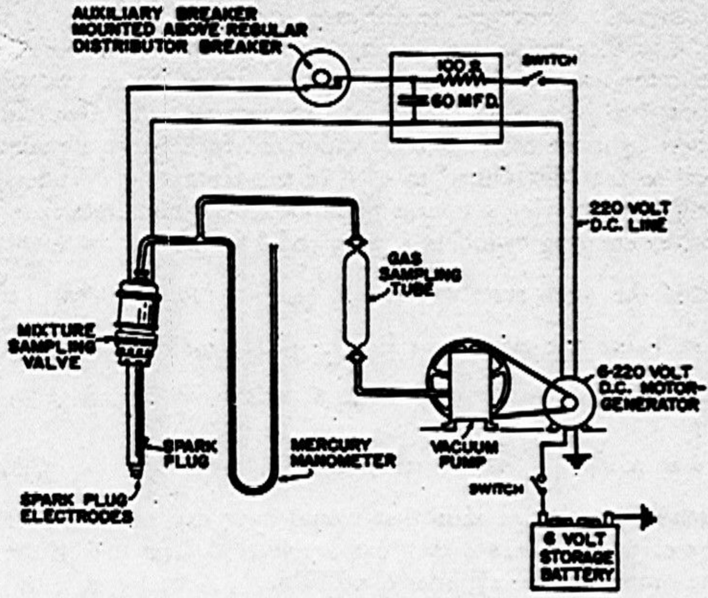

FIG. 2 DIAGRAMMATIC SKETCH OF MIXTURE SAMPLING VALVE AND ASSOCIATED EQUIPMENT.

The remainder of the heavy liquid portion is distributed unequally to the cylinders, the quantity entering any given cylinder depending on the shape, cross-sectional area and inclination of the manifold, engine speed, surging of the mixture within the manifold, and other factors. Thus, in the three typical cases, one or more cylinders may receive substantially only air and the lighter ends which leave the manifold in the vapor state; others may receive air, vaporized light ends and liquid heavy ends in approximately the same air-fuel ratio as that of the mixture leaving the carburetor, and still others may receive air, vaporized light ends and liquid heavy ends in the same proportions as in the second case plus the liquid heavy ends which, under conditions of perfect distribution, would have entered the first-mentioned cylinder or group of cylinders. Other cylinders may receive liquid heavy ends in any amounts between the two limits just described, and it would be possible for one or more cylinders to receive even more liquid heavy ends than the richest cylinder cited. The three typical distribution conditions are summarized in Table 3 in which the fuel entering the carburetor is assumed to contain *L* lb. of light ends, which leave the manifold in the vapor state, for each *H* lb. of heavy ends that leave the manifold in the liquid state.

Table 3 – Three Typical Fuel Distribution Conditions in the Multicylinder Engine

Cylinder Type	Air Received by Cylinder, lb.	Vaporized Light Ends Received by Cylinder, lb.	Liquid Heavy Ends Received by Cylinder, lb.	Cylinder Air-fuel Ratio	Carburetor Air-fuel Ratio	Cylinder Mixture Strength Relative to Carburetor
A	M	L	O	$\frac{M}{L}$	$\frac{M}{L+H}$	Leaner
B	M	L	H	$\frac{M}{L+H}$	$\frac{M}{L+H}$	Equal
C	M	L	2 H	$\frac{M}{L+2H}$	$\frac{M}{L+H}$	Richer

Mean air-fuel ratio — carburetor air-fuel ratio

$$= \frac{3}{\frac{L}{M} + \frac{L+H}{M} + \frac{L+2H}{M}} = \frac{M}{L+H}$$

Assume, for example, in the case of cylinder group 1, 2, 7, and 8 of the engine in Table 2, that the average cylinder air-fuel ratio was equal to the carburetor air-fuel ratio, that no liquid fuel was distributed by entrainment, and that the leanest cylinder received only vaporized fuel, in an amount equal to that distributed to each of the remaining cylinders, along with varying amounts of liquid fuel. The fraction of the fuel entering cylinders 1, 2, 7, and 8 which left the manifold in the vapor state was $100 \times \frac{14.4}{16.01} = 90.0$ per cent. If M in Table 3 is taken as 1 lb. $(L + H)$ for cylinders 1, 2, 7, and 8 was $\frac{1}{14.4} = 0.0695$ lb. L was $\frac{1}{16.01} = 0.0625$ lb. H was $0.0695 - 0.0625 = 0.0070$ lb. $\frac{M}{L + 2H} = 13.08$, which is the air-fuel ratio that would have existed in one or more of the cylinders if they had received air and fuel in the same proportions as cylinder C of Table 3. Actually no cylinder in this group received such a mixture, but cylinder 8 received a mixture having an air-fuel ratio of 12.37, or 0.0809 lb. fuel per lb. of air, consisting of L lb. of vaporized light ends and 2.63 H lb. of liquid heavy ends.

Effect of Variable Fuel Distribution on Knocking

The non-uniform distribution of fuel to the cylinders as just described gives rise to two conditions which are of vital importance in the fuel-knocking problem, namely:

(1) The air-fuel ratios of the mixtures in the cylinders may vary widely and cause a substantial variation in knocking tendency among the cylinders.

(2) The composition of the fuel in the cylinders may be quite different and cause a variation in knocking tendency in addition to that introduced by the differences in air-fuel ratios.

Because of these conditions, independent of all others, there is ample cause for a marked variation in the knocking tendency of the fuel in the individual cylinders of an engine operated under conditions of imperfect distribution. Whether the cylinders of Type A, B or C (Table 3) have the greatest knocking tendency depends on the following three factors:

(1) The carburetor air-fuel ratio.

(2) The degree of vaporization and hence the magnitude of the variation in cylinder air-fuel ratio.

(3) The relative concentration of antiknock value in the vaporized light ends and the unvaporized heavy ends of the fuel.

Adaptation of the Single-Cylinder Engine

The laboratory knock-testing engine, operated in the conventional manner, permits no prediction of the behavior of a fuel under variations of the three conditions just enumerated. Under such a procedure it is useful only for the evaluation of the knocking tendency of a fuel under a range of cylinder conditions when the composition and air-fuel ratio of the mixture entering the cylinder are the same as for the mixture leaving the carburetor.

A fundamental study on a multicylinder engine of fuel distribution and its effect on knocking is surrounded by many difficulties. The richness of the mixture which enters the several cylinders can be determined most conveniently by exhaust-gas analysis as described previously. The knocking tendency of the fuel in the individual cylinders is not measurable by any instrumentation developed to date but can be evaluated by audibility methods to be described later. The maintenance of constant conditions of operation is difficult because of uncontrollable variations in ignition timing and in temperatures. It is, therefore, highly desirable to adapt the C.F.R. engine to the study of this problem, inasmuch as the above-mentioned variations in operating conditions have been minimized.

Fig. 3 shows four intake manifolds for the C.F.R. engine which have been designed to permit a study, under controlled conditions, of the effects on knocking caused by variations of fuel distribution in the multicylinder engine. In the arrangement shown in Fig. 3*a* (upper left), a short spacer bolted directly to the engine cylinder is fitted with a thermometer for the indication of mixture temperature. A standard C.F.R. manifold, with its mixture heater, is bolted to the spacer, the flange normally bolted to the carburetor being attached to the spacer. A specially designed water-jacketed manifold section is provided with two flanges, one of which is connected to the standard C.F.R. manifold and the other to the standard C.F.R. carburetor body.

After the fuel-air mixture leaves the carburetor, it is forced to flow down through the jacketed section of manifold and up through the standard C.F.R. manifold before entering the cylinder. Unvaporized fuel which is not carried into the cylinder by entrainment in the air stream drains from the jacketed section of the manifold into a graduated burette. The degree of fuel vaporization is controlled by the temperature of the water that enters the manifold jacket. Water is heated or cooled externally and mixed with tap water to produce the temperature required to effect the desired degree of vaporization. Any final mixture temperature higher than that existing in the jacketed manifold may be obtained by the use of the standard C.F.R. mixture heater.

In order that the rate of carburetor fuel consumption may be measured, the standard C.F.R. integral float-bowl and fuel-container assemblies are replaced by separate float-bowl and fuel-container assemblies from the Series 30 engine. Three-way cocks and graduated glass bulbs permit the volumetric measurement of fuel entering the carburetor. An air-conditioning unit, as described in the June, 1937, issue of the S.A.E. JOURNAL, page 250,[2] maintains carburetor air at constant temperature and humidity and provides for air measurement by means of a round-edge orifice similar to that described in the 1936 Transactions of the S.A.E., p. 97.[3]

The arrangement shown in Fig. 3*b* (upper right) has been found to effect a more complete separation of liquid fuel from the mixture of air and vaporized fuel and, thus, to simulate more closely the functioning of the multicylinder engine manifold. Essentially it is the set-up shown in Fig. 3*a* with the regular C.F.R. manifold and special jacketed section reversed in position. Variation in the position of the piston in the surge cylinder attached to the liquid fuel drain at the bottom of the manifold permits, with little or no application of heat to the manifold, a wide range of adjustment of the fraction of the fuel drained from the manifold in the liquid state. It is impossible, however, to obtain with this manifold a final mixture temperature higher than that of the liquid in the jacket. This restriction prevents operation with cylinder conditions having a severity much in excess of those for the Research Method.

The manifold shown in Fig. 3*c* (lower left) permits the use of any desired final mixture temperature higher than approximately 200 deg. fahr., effects as good separation of vaporized and liquid fuel as the manifold shown in Fig. 3*b*, and requires no surge chamber. The separating chamber has a cross-sectional area over four times as great as that of the remainder of the manifold and consequently reduces the velocity of flow to a point that ensures good separation of liquid fuel from the stream of air and vaporized fuel. Heat may be applied to the mixture by conventional immersion

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[2] See S.A.E. TRANSACTIONS, June, 1937, p. 250: Earl Bartholomew's discussion of "Influence of Humidity on Knock Ratings," by J. R. MacGregor.

[3] See S.A.E. TRANSACTIONS, March, 1936, p. 97: Earl Bartholomew's discussion of "Relation of Exhaust-Gas Composition to Air-Fuel Ratio," by B. A. D'Alleva and W. G. Lovell.

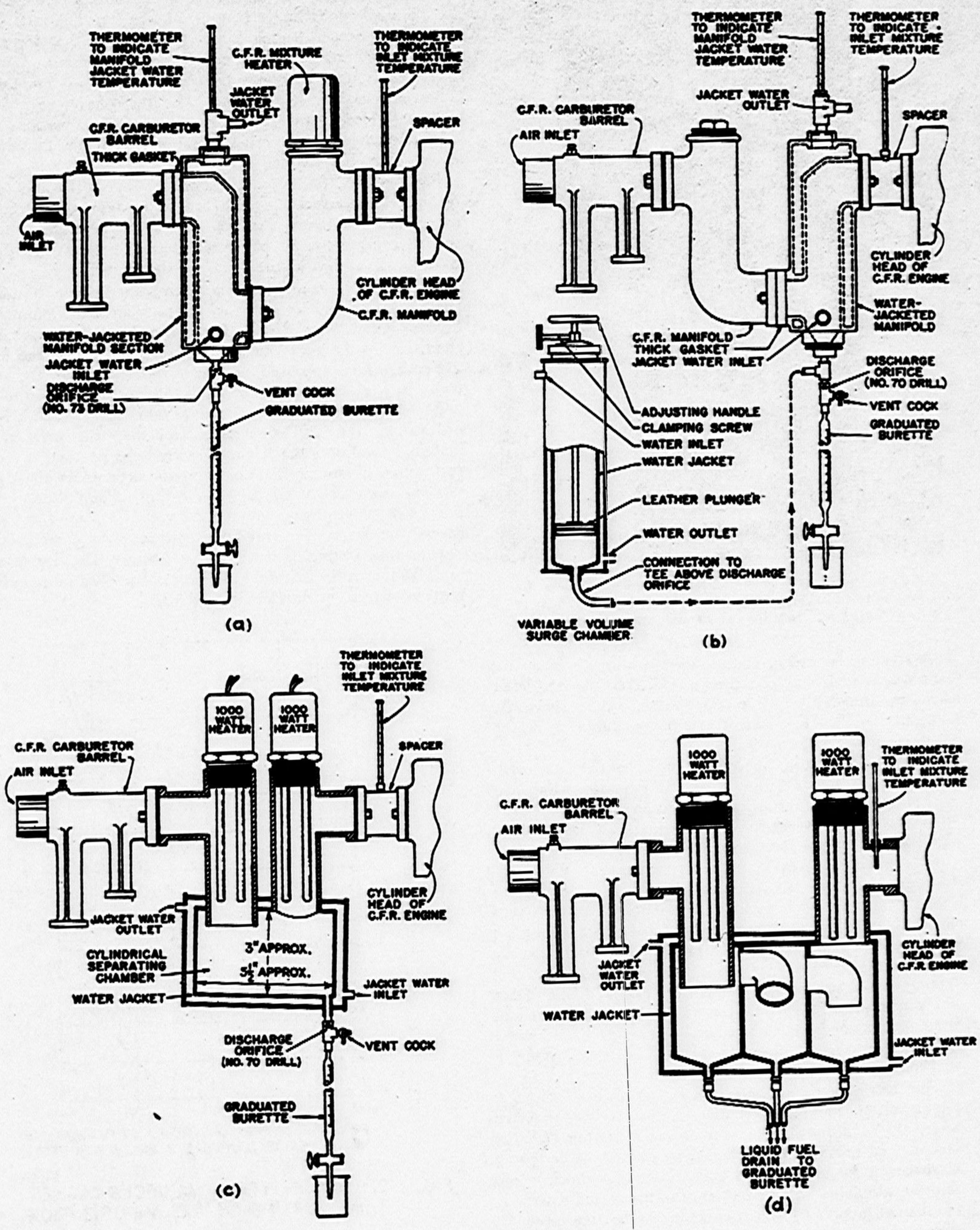
THERMOMETER TO INDICATE MANIFOLD JACKET WATER TEMPERATURE
C.F.R. MIXTURE HEATER
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
JACKET WATER OUTLET
C.F.R. CARBURETOR BARREL
THICK GASKET
SPACER
AIR INLET
CYLINDER HEAD OF C.F.R. ENGINE
C.F.R. MANIFOLD
WATER-JACKETED MANIFOLD SECTION
JACKET WATER INLET
DISCHARGE ORIFICE (NO. 73 DRILL)
VENT COCK
GRADUATED BURETTE
(a)
THERMOMETER TO INDICATE MANIFOLD JACKET WATER TEMPERATURE
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
JACKET WATER OUTLET
C.F.R. CARBURETOR BARREL
AIR INLET
SPACER
CYLINDER HEAD OF C.F.R. ENGINE
WATER-JACKETED MANIFOLD
C.F.R. MANIFOLD
THICK GASKET
JACKET WATER INLET
DISCHARGE ORIFICE (NO. 70 DRILL)
VENT COCK
GRADUATED BURETTE
ADJUSTING HANDLE
CLAMPING SCREW
WATER INLET
WATER JACKET
LEATHER PLUNGER
WATER OUTLET
CONNECTION TO TEE ABOVE DISCHARGE ORIFICE
VARIABLE VOLUME SURGE CHAMBER
(b)
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
1000 WATT HEATER
1000 WATT HEATER
C.F.R. CARBURETOR BARREL
AIR INLET
SPACER
CYLINDER HEAD OF C.F.R. ENGINE
JACKET WATER OUTLET
CYLINDRICAL SEPARATING CHAMBER
3" APPROX.
5½" APPROX.
WATER JACKET
JACKET WATER INLET
DISCHARGE ORIFICE (NO. 70 DRILL)
VENT COCK
GRADUATED BURETTE
(c)
1000 WATT HEATER
1000 WATT HEATER
THERMOMETER TO INDICATE INLET MIXTURE TEMPERATURE
C.F.R. CARBURETOR BARREL
AIR INLET
CYLINDER HEAD OF C.F.R. ENGINE
JACKET WATER OUTLET
WATER JACKET
JACKET WATER INLET
LIQUID FUEL DRAIN TO GRADUATED BURETTE
(d)

FIG.3 DIAGRAMMATIC SKETCHES OF INTAKE MANIFOLDS FOR THE C.F.R. ENGINE WHICH SIMULATE THE EFFECTS OF IMPERFECT MIXTURE DISTRIBUTION IN THE MULTICYLINDER ENGINE.

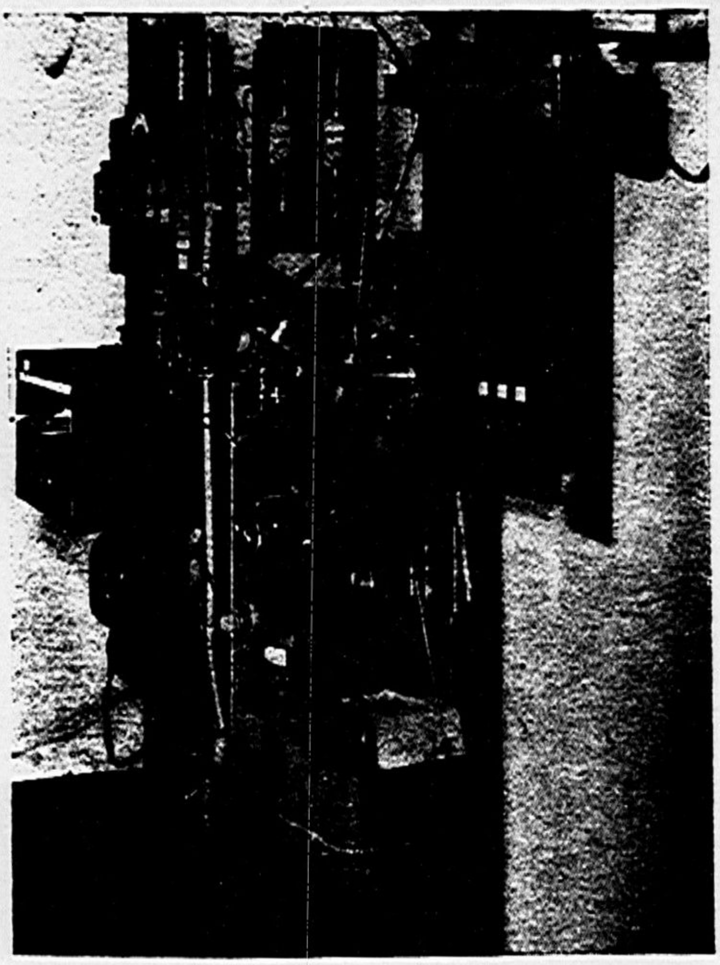

Fig. 4 – Modified Intake Manifold and Fuel System Installed on the C.F.R. Engine for Studies of the Effects of Imperfect Fuel Distribution in the Automobile Engine

heaters before or after it enters the separating chamber and the chamber itself may be heated or cooled by the circulation of water through the surrounding jacket. Fig. 4 shows the complete modified fuel system and the manifold of Fig. 3*c* installed on a C.F.R. engine.

The manifold of Fig. 3*d* (lower right) is of the same essential design as that of Fig. 3*c* but is provided with three separating chambers instead of one. Experience to date with this design is insufficient to indicate whether it possesses any significant advantage over that shown in Fig. 3*c*.

Since provision is made for the measurement of air and fuel delivered to the carburetor and the unvaporized fuel bled from the manifold, it is possible to compute the air-fuel ratios of the mixtures leaving the carburetor and of those entering the cylinder. When unvaporized fuel is withdrawn from the manifold, the mixture entering the cylinder is leaner than that leaving the carburetor and the distribution condition is that of Cylinder *A* in Table 3. When no unvaporized fuel is bled from the manifold, the cylinder and carburetor air-fuel ratios are equal and the distribution condition is that of Cylinder *B* in Table 3.

The distribution condition of Cylinder *C* in Table 3 is duplicated on the single-cylinder engine by running first under the conditions of Cylinder *A* and collecting *H* lb. of unvaporized heavy ends. It would be difficult to design an arrangement for the modified C.F.R. manifold which would feed the additional liquid heavy ends to the normal charge in the desired ratio. It is easy to accomplish the same result by adding the excess heavy ends for Cylinder *C* directly to the original fuel in the ratio of *H* lb. of heavy ends for each $(L + H)$ lb. of original fuel and feeding this mixture to the carburetor. The carburetor air-fuel ratio is adjusted to $\frac{L+H}{L+2H} \times$ (air-fuel ratio for Cylinder *B*) $= \frac{M}{L+2H}$, and the manifold heating is adjusted for complete vaporization. It should be noted that, during the determination of the data for the curve of Cylinder *C*, the carburetor air-fuel ratio for the single-cylinder engine is not the same as that for the multicylinder engine whose performance is simulated.

The ratio of *L* to *H* may be varied over a wide range by adjustment of manifold heating. The liquid heavy ends also may be added to the original fuel in other proportions to duplicate other mixture ratios found to exist in the cylinders of an automobile engine.

Fig. 5 shows, for 1937 C.F.R. winter fuel WB-2 and secondary reference fuels A-4 and C-10 the relation of the degree of vaporization in the manifold to the temperature of the water leaving the jacket of the manifold of Fig. 3*a*. The distillation curves for the three fuels are shown in Fig. 6.

The degree of vaporization in the modified single cylinder engine manifold varies with fuel volatility as in the manifold of the multicylinder engine. Fig. 7 shows for one manifold condition for the 1937 C.F.R. preferred winter fuels, and for 3 blends of the approved secondary reference fuels, the relation between the per cent of fuel vaporized in the manifold and the per cent over at 310 deg. fahr. in the A.S.T.M. distillation, whereas Fig. 8 shows, for the same fuels and manifold condition, the relation between the per cent of the fuel vaporized in the manifold and the area under the distillation curve above 80 deg. fahr. Figs. 7 and 8 show a fairly consistent relation between the variables plotted but, of course, the degree of vaporization depends also on the latent heat of vaporization of the fuel components. The degree of vaporization in the manifold might correlate even more satisfactorily with data from an air distillation.

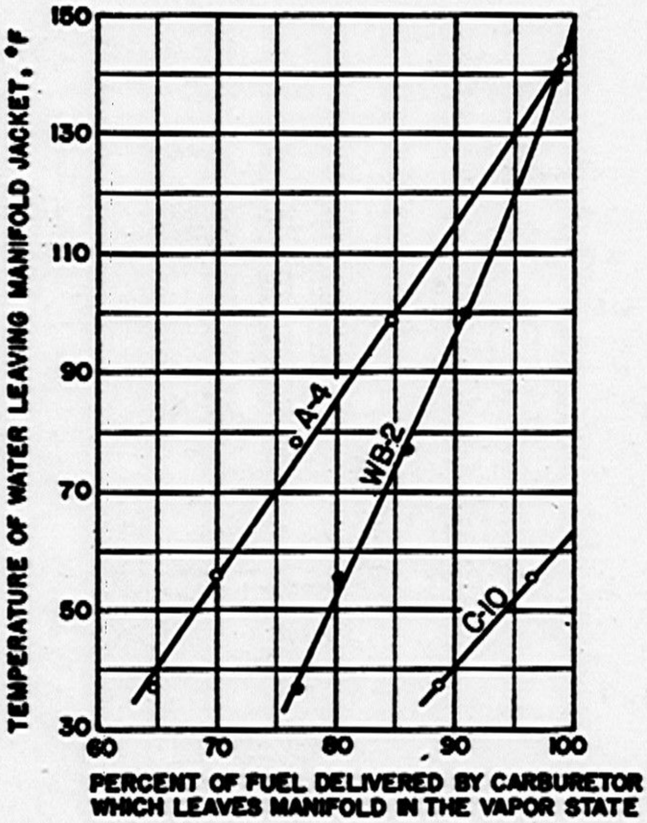

FIG. 5. EFFECT OF INTAKE MANIFOLD JACKET TEMPERATURE ON FUEL VAPORIZATION

C.F.R. ENGINE, SPECIAL MANIFOLD
AND CARBURETOR (FIG. 3a)
FIXED CARBURETOR SETTING
MIXTURE TEMP. ENTERING CYLINDER, 300° F.
SPARK ADVANCE, 21°
CYLINDER JACKET TEMP., 212° F.
ENGINE SPEED, 900 R.P.M.

Knock Ratings with the Bled Manifold

After the carburetor float level and manifold jacket temperature have been adjusted to give the desired carburetor and cylinder air-fuel ratios respectively, and the final mixture temperature has been brought to the desired figure, the general procedure for making knock ratings with the modified manifold is the same as for the Motor Method. The compression ratio is adjusted for standard knock intensity and the bouncing-pin indicator is used to measure knocking tendency.

If the primary reference fuels are used, the fuel octane numbers obtained are likely to differ considerably, under conditions of partial fuel vaporization, from those obtained by the use of the approved secondary reference fuels. Within the range of manifold conditions of practical interest, the vaporization of heptane-octane mixtures is substantially complete, hence octane numbers obtained by the use of the primary reference fuels have the same meaning as those obtained by the Motor or Research Methods or by any other method which employs the conventional manifold. On the other hand, the volatility of the C-10 and A-5 reference fuels is quite different; therefore, under any condition of partial vaporization, a larger fraction of the C-10 component is vaporized in the manifold. As the concentration of A-5 in the blend is increased, the per cent of the blend leaving the manifold in the vapor state is decreased, and the inherent antiknock value of the reference-fuel mixture which enters the cylinder decreases less rapidly than does the per cent of C-10 in the blend. The result is that, under these conditions, a given increment of antiknock value matched against blends of the secondary reference fuels is interpreted as representing a larger difference in octane numbers than when matched directly against the primary reference fuels. The same condition presumably exists during road knock tests and raises the question of the possible relation between it and the occasionally observed relative insensitivity of cars to changes in the per cent of C-10 in the blends of the secondary reference fuels.

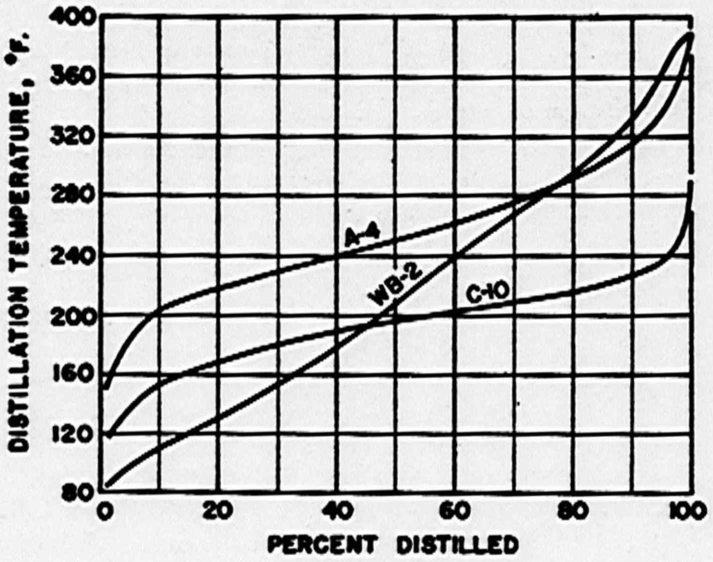

FIG. 6 DISTILLATION CURVES OF FUELS WHOSE VAPORIZATION IN THE MANIFOLD IS ILLUSTRATED IN FIG. 5.

When a measure of the difference in knocking tendency of fuels under conditions of imperfect distribution is desired in units bearing a definite relation to critical compression ratio, it is customary to make the comparisons directly against heptane-octane mixtures. When the problem is to correlate laboratory ratings with those obtained on the road with the secondary reference fuels, the latter reference fuels are used in the laboratory. If the conditions of engine operation on the road are duplicated in the laboratory, the fractions of the reference-fuel blends delivered by the carburetor that leave the manifold in the vapor state are the same in both cases. If the octane numbers in terms of the primary reference fuels are to be determined at one time for one or more fuels at several degrees of imperfect vaporization, it is customary to determine the compression ratio for each that produces a knockmeter reading of 50 and to read octane numbers from a curve of octane number versus compression ratio for constant knock intensity, made at the conclusion of the tests.

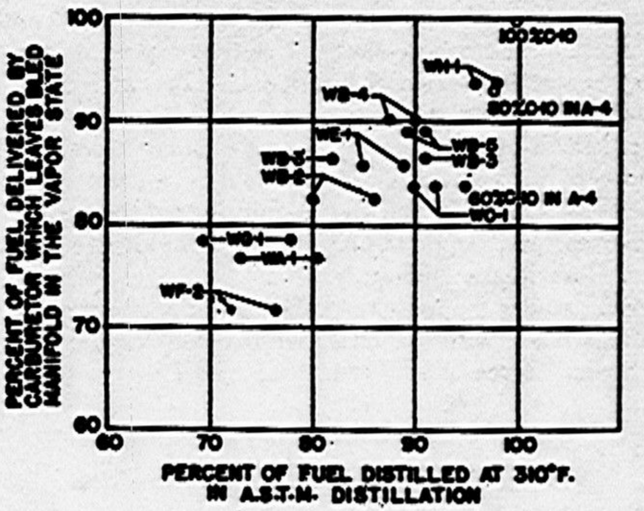

FIG. 7 RELATION BETWEEN THE PERCENT OF FUEL DELIVERED BY THE CARBURETOR WHICH LEAVES THE BLED MANIFOLD IN THE VAPOR STATE, AND THE PERCENT OVER AT 310 °F. IN THE A.S.T.M. DISTILLATION.

C.F.R. ENGINE, SPECIAL MANIFOLD
AND CARBURETOR (FIG. 3a)
MAXIMUM KNOCK AIR-FUEL RATIO
MANIFOLD JACKET TEMP., 68°F.
CARBURETOR AIR TEMP., 80°F.
MIXTURE TEMP. ENTERING CYLINDER, 85°F.
SPARK ADVANCE, MOTOR METHOD
CYLINDER JACKET TEMP., 212°F.
ENGINE SPEED, 900 R.P.M.

When the degree of vaporization is low, as for example 50 to 60 per cent, the unsteady state following a change from one fuel to another is of quite long duration. In any case

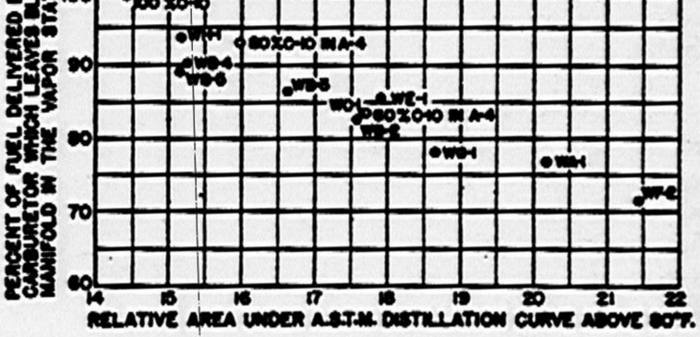

FIG. 8 RELATION BETWEEN THE PERCENT OF FUEL DELIVERED BY THE CARBURETOR WHICH LEAVES THE BLED MANIFOLD IN THE VAPOR STATE, AND THE RELATIVE AREA UNDER THE A.S.T.M. DISTILLATION CURVE

C.F.R. ENGINE, SPECIAL MANIFOLD
AND CARBURETOR (FIG. 3a)
MAXIMUM KNOCK AIR-FUEL RATIO
MANIFOLD JACKET TEMP., 68°F.
CARBURETOR AIR TEMP., 80°F.
MIXTURE TEMP. ENTERING CYLINDER, 85°F.
SPARK ADVANCE, MOTOR METHOD
CYLINDER JACKET TEMP., 212°F.
ENGINE SPEED, 900 R.P.M.

where vaporization is not perfect, the unsteady state persists for a longer period than under the Motor Method conditions. There appears to be no way at constant speed of the laboratory engine to shorten the period of transient conditions. In tests on the road the car engine is usually operated at high speed for a brief period, following a change from one fuel to another, during which the manifold is washed clean of the previous fuel more quickly. A multi-speed, power-absorption generator and larger samples of fuel should permit a similar procedure in the laboratory.

Even under the most favorable conditions in the laboratory, ratings made with the bled manifold require more time and a higher degree of skill than those made by the conventional methods. It is believed that reproducibility of ratings is about the same. Complete studies of fuels under a wide range of carburetor and manifold conditions, carried out by the procedure just described, should constitute a valuable tool for both the fuel refiner and automotive engineer.

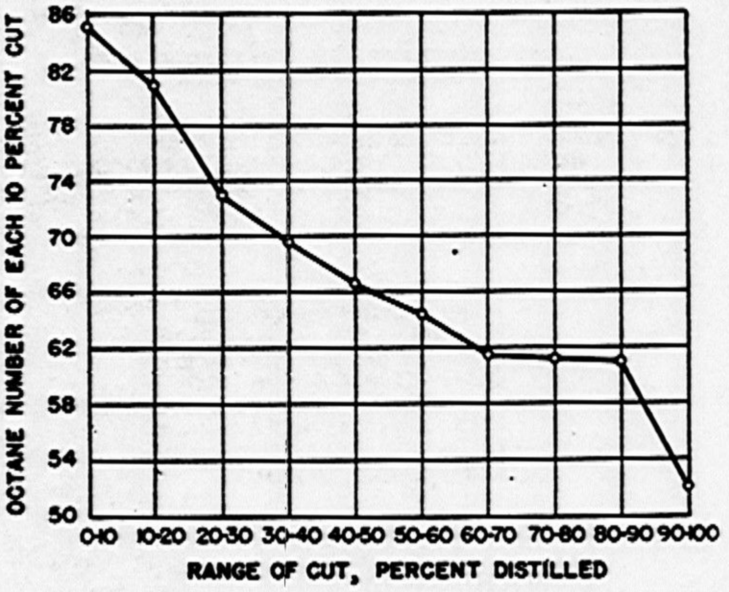

FIG. 9 ANTIKNOCK VALUE OF TEN EQUAL CUTS OF A TYPICAL COMMERCIAL BASE GASOLINE SEPARATED BY SIMPLE DISTILLATION.

C.F.R. ENGINE, STANDARD MANIFOLD
NO MIXTURE HEATER
MAXIMUM KNOCK AIR-FUEL RATIO
MIXTURE TEMP. ENTERING CYLINDER, 140°F.
(OBTAINED BY HEATING INTAKE AIR)
SPARK ADVANCE, 18°
CYLINDER JACKET TEMP., 212°F.
ENGINE SPEED, 600 R.P.M.

The result of tests made on a single-cylinder engine equipped with the bled manifold might be used as a basis for the correction of ratings obtained by the conventional procedure. It would be preferable to obtain final ratings, by a single engine test, which would be corrected for the distribution effects in the car engine and in agreement with the average ratings obtained in cars on the road. Possibly a set of engine conditions, including a modification of the intake manifold, could be found to produce such ratings.

Distribution through the Fuel Distillation Range

It has been stated previously that the relative knocking tendency of the lean, normal, and rich cylinders (denoted by *A*, *B*, and *C* respectively in Table 3) depends on the carburetor air-fuel ratio, the degree of vaporization in the carburetor and manifold, and the relative concentration of antiknock value in the vaporized light ends and unvaporized heavy ends of the fuel. The distribution of antiknock value through the distillation range may be measured by separating the fuel into a number of cuts by distillation and determining the octane number of each by any of the conventional knock-testing methods. Fig. 9 shows such data for a typical commercial lead-free gasoline base stock so treated. The most volatile fractions have the highest antiknock value, and the heavier cuts have progressively lower octane number.

Such a procedure is time-consuming and not required for the determination of the distribution of antiknock value within the practical limits of interest. A more convenient method is to effect the distillation and measure antiknock value simultaneously in the engine itself. Fig. 10 shows the octane numbers of a number of cuts of the 1937 C.F.R. preferred winter road-test fuels and of five other fuels. The cuts were made by varying the degree of vaporization in the manifold and, in each case, they included the most volatile fractions of the fuel. All ratings were made at the air-fuel ratio for maximum knock. The curves are stopped at approximately the 50 per cent vaporization point for the least volatile fuels, inasmuch as this point appears to represent roughly the extreme condition to be met on the road.

Fuels may be separated roughly into three broad classes in accordance with the distribution of antiknock value along their distillation curves, as follows:

(1) Those whose cuts by simple distillation from the 50 to 100 per cent points have approximately the same antiknock value as the first 50 per cent cut as, for example, fuel WA-1 in Fig. 10*a*.

(2) Those whose cuts by simple distillation from the 50 to 100 per cent points have constantly increasing antiknock value, illustrated by specially blended fuel S-6 in Fig. 10*a*.

(3) Those whose cuts between the 50 and 100 per cent points have constantly decreasing antiknock value, such as fuel WG-1 of Fig. 10*b*.

Among commercial lead-free fuels, those of the third class are by far the most numerous; however, the degree of concentration of antiknock value in the light ends, although variable, never approaches that of specially blended fuel S-11 referred to herein (Fig. 10*b*). Commercial lead-free fuels of the first class are encountered occasionally and fuels of the third class may be changed, by the addition of lead tetraethyl, to have the characteristics of those of the first class. Those in the second class are not available commercially but may be made for research purposes by careful blending of selected components.

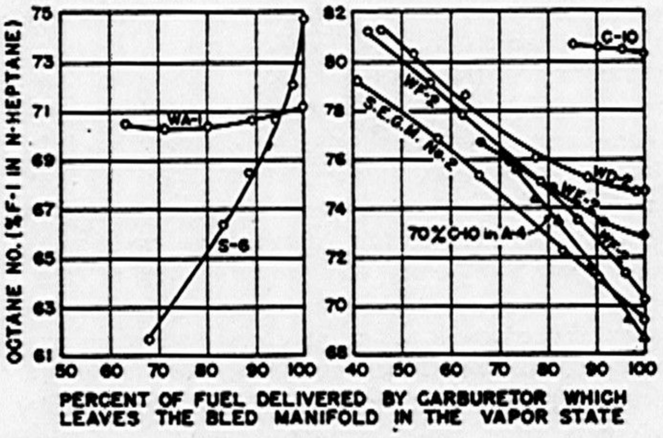

FIG. 10a VARIATION OF THE ANTIKNOCK VALUE OF THE VAPORIZED PORTION OF 4 OF THE 1937 C.F.R. WINTER FUELS AND 4 OTHER FUELS WITH THE DEGREE OF VAPORIZATION.

C.F.R. ENGINE, SPECIAL MANIFOLD (FIG. 3c).
CYLINDER AIR-FUEL RATIO FOR
MAXIMUM KNOCK.
TEMP. MIXTURE ENTERING CYLINDER, 220°F.
SPARK ADVANCE, 18°
ENGINE SPEED, 600 R.P.M.

Variation of Cylinder Air-Fuel Ratio

It has been pointed out in Table 3 that imperfect distribution causes some cylinders of the multicylinder engine to receive mixtures richer and leaner respectively than that leaving the carburetor, whereas others may receive a mixture having substantially the same composition and air-fuel ratio as that leaving the carburetor. Inasmuch as the knocking tendency of the fuel in the cylinder is influenced markedly by air-fuel ratio as well as by the composition of the fuel, it is interesting to investigate the combined effect of these two variables resulting from imperfect distribution of the fuel in the multicylinder engine. It is possible to predict for any given fuel the relative knocking tendency of the several cylinders by the construction of any one of a number of different charts after a series of tests of the fuel made on a C.F.R. engine equipped with one of the manifolds shown in Fig. 3. The type of chart which is most satisfactory depends on the information desired.

Inasmuch as the most frequent problem is the prediction of the relative knocking tendency of a group of fuels in a single car under a given set of conditions, the most valuable type of chart is usually one constructed on the basis of constant carburetor air-fuel ratio. The chart of Fig. 11 is constructed at a carburetor air-fuel ratio of 11.21 for fuel S.E.G.M. No. 2, which is a typical commercial, regular grade, lead-free base gasoline comprising a mixture of straight-run and cracked stocks. The scale of ordinates is critical compression ratio, defined as the compression ratio required for a knockmeter reading of 50. The scale of abscissae is the per cent of the fuel delivered by the carburetor which leaves the manifold in the vapor state.

Data for the construction of the curve for Cylinder *A* are obtained by operating the engine at the desired speed, spark advance, and final mixture temperature and, by adjustment of manifold temperatures, varying the per cent of the fuel delivered by the carburetor which is bled from the manifold in the liquid state. The compression ratio for a knockmeter reading of 50 is determined for each rate of fuel bleeding and the curve is drawn through the points so determined. If the air and fuel passing through the carburetor and the liquid fuel bled from the manifold are measured, it is possible to compute the air-fuel ratio of the mixture which enters the cylinder. This information is shown beside some of the data points in Fig. 11.

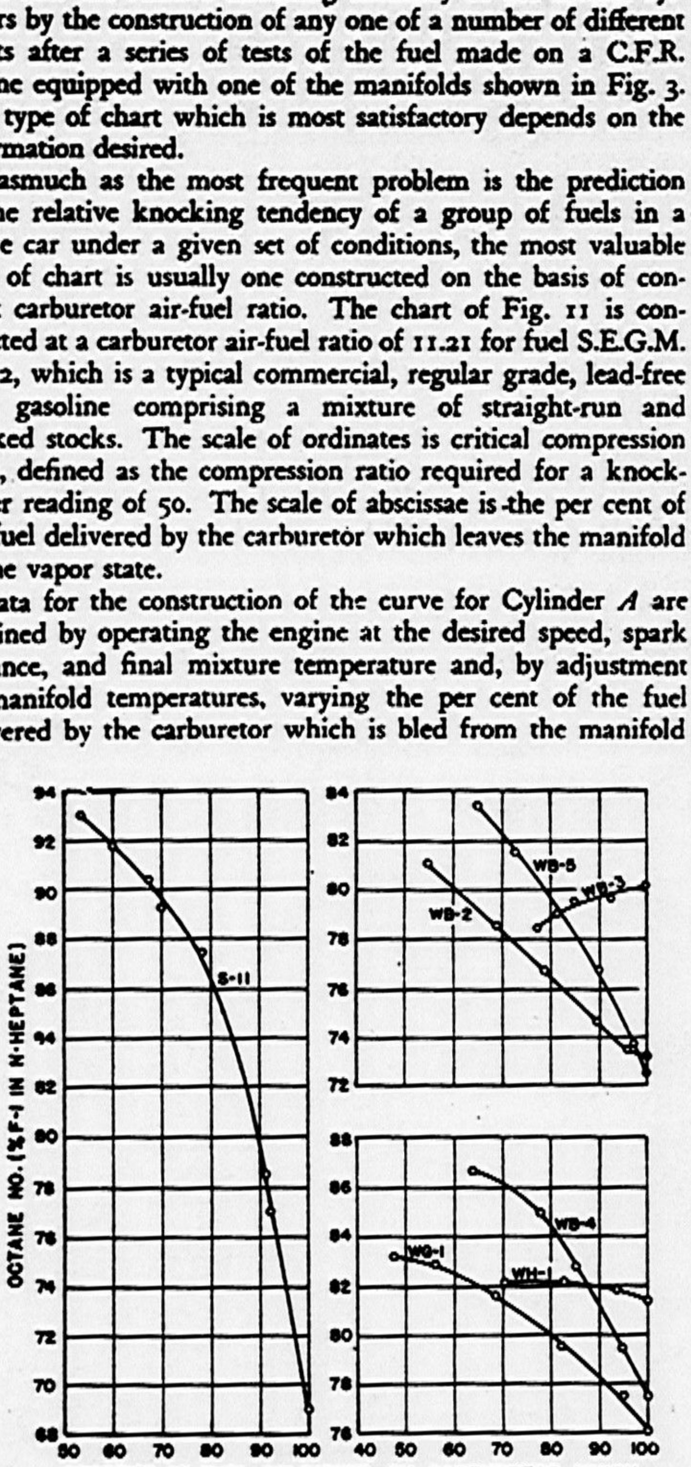

FIG. 10b VARIATION OF THE ANTIKNOCK VALUE OF THE VAPORIZED PORTION OF 6 OF THE 1937 C.F.R. WINTER FUELS AND 1 OTHER FUEL WITH THE DEGREE OF VAPORIZATION.

C.F.R. ENGINE, SPECIAL MANIFOLD (FIG. 3a).
CYLINDER AIR-FUEL RATIO FOR MAXIMUM KNOCK.
TEMP. MIXTURE ENTERING CYLINDER, 220 °F.
SPARK ADVANCE, 18°
ENGINE SPEED, 600 R.P.M.

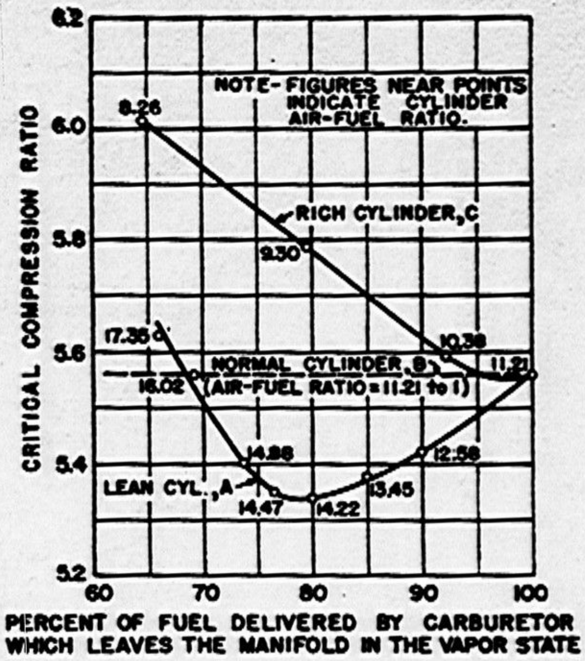

FIG. 11 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

TYPICAL COMMERCIAL LEAD-FREE BASE STOCK.
AIR-FUEL RATIO AT THE CARBURETOR, 11.21

The air-fuel ratio and composition of the mixture in Cylinder *A* under the condition of complete fuel vaporization would apply to the normal Cylinder *B* of the multicylinder engine regardless of the degree of fuel vaporization in the manifold. The curve for Cylinder *B* therefore is a straight line, parallel to the horizontal axis, and intersecting the curve for Cylinder *A* at the 100 per cent vaporization point.

It will be recalled that Cylinder *C* of Table 3 receives a normal charge plus the liquid heavy ends which, under conditions of perfect fuel distribution, would have entered Cylinder *A*. Data for the construction of the curve for Cylinder *C* are determined in the manner previously described. When vaporization is complete in the manifold, *H* becomes zero and the curve for Cylinder *C* has a point in common with the curves for Cylinders *A* and *B*.

Fig. 10*a* shows that the lighter fractions of fuel S.E.G.M. No. 2 have the higher antiknock value, nevertheless, Fig. 11 indicates that, under conditions of imperfect fuel distribution at a carburetor air-fuel ratio of 11.21, Cylinder *A* which receives the fuel of highest inherent antiknock value has the greatest knocking tendency of the three cylinders when 69 to 100 per cent of the fuel leaves the manifold in the vapor state. Furthermore, Cylinder *C*, which receives fuel having the lowest inherent antiknock value, has the least tendency to knock. As the degree of vaporization decreases, the air-fuel ratio of Cylinder *A* decreases from 11.21 toward the maximum knock air-fuel ratio, and this change in air-fuel ratio has a greater influence on the knocking tendency of the fuel in the cylinder than the increase in the inherent antiknock value of the fuel fraction itself. Similarly, the richening of the mixture in Cylinder *C* under the same conditions has a

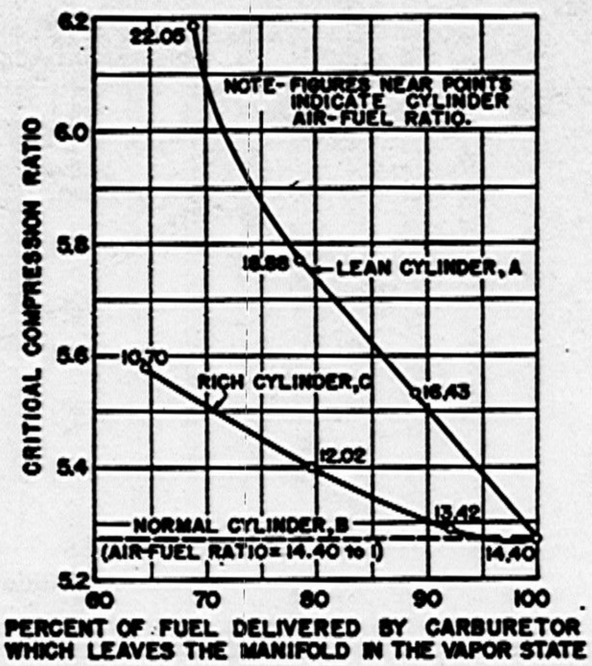

FIG. 12 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

TYPICAL COMMERCIAL LEAD-FREE BASE STOCK.
AIR-FUEL RATIO AT THE CARBURETOR, 14.40
(MAXIMUM KNOCK)

greater effect on the knocking tendency of the fuel in this cylinder than the decrease in the inherent antiknock value of the fuel.

The base fuel and conditions illustrated in Fig. 11 are those of commercial interest. As a matter of academic interest and to provide a more complete background for the fuel-distribution problem, Figs. 12 to 16 have been prepared. Fig. 12 is constructed for fuel S.E.G.M. No. 2 illustrated in Fig. 11 but at the carburetor air-fuel ratio for maximum knock. A full-throttle setting so lean is not used in cars because of the resultant loss of power at all speeds and uneven operation at low speeds. The carburetor conditions illustrated in Figs. 13 and 15 are in the normal range but the fuels for these figures, as well as for Figs. 14 and 16, are specially blended extreme types which are not representative of commercial gasolines.

Fig. 12 is similar to Fig. 11 in all respects except that the carburetor air-fuel ratio is the maximum knock ratio for fuel S.E.G.M. No. 2. At this carburetor setting the air-fuel ratio for Cylinder *A* becomes leaner than that for maximum knock as the degree of fuel vaporization is decreased. Simultaneously the inherent antiknock value of the fuel in the cylinder increases. The result is a marked rate of increase in critical compression ratio as the degree of vaporization decreases.

Under the same conditions, the air-fuel ratio of the mixture in Cylinder *C* becomes richer than that for maximum knock and for this reason the fuel tends to knock less. At the same time, however, the inherent antiknock value of the fuel decreases. The result of these two opposed tendencies is that, as the degree of vaporization decreases, the knocking tendency of Cylinder *C* at first remains close to that of normal Cylinder *B*, but becomes less than that for Cylinder *B* as the degree of vaporization is decreased still further.

The differential in antiknock value between the light and heavy ends of fuel S.E.G.M. No. 2 is within the normal range for commercial fuels. Fuel S-11 was specially blended to have light and heavy ends of extremely high and low antiknock value respectively. This abnormal distribution of antiknock value through the fuel fractions reverses the relative knocking tendency of the rich and lean cylinders. Fig. 13 shows for this fuel the relative knocking tendency of the three typical cylinders, *A*, *B*, and *C*, when the carburetor air-fuel ratio is 11.46. The difference between the knocking tendency of the light and heavy fractions of this fuel is so great that, as the air-fuel ratio of Cylinder *A* moves from 11.46 toward the maximum knock air-fuel ratio, the increase in inherent antiknock value of the fuel in the cylinder almost exactly balances the effect of the change in cylinder air-fuel ratio. At cylinder air-fuel ratios leaner than for maximum knock, both influences tend to decrease knocking and the curve for Cylinder *A* rises rapidly.

As the degree of vaporization is decreased, the knock-suppressing effect of the richer mixture in Cylinder *C* is not sufficient to compensate for the depressing effect of the additional heavy ends of very low antiknock value. Rich Cylinder *C*, therefore, has the greatest knocking tendency, relative to that of Cylinders *A* and *B*, when vaporization is between 71 and 100 per cent.

When the carburetor air-fuel ratio is that for maximum knock (Fig. 14), the rich Cylinder *C* still has the greatest knocking tendency, relative to Cylinders *A* and *B*, between vaporization rates of 72 and 100 per cent. Fig. 14 shows, however, that its knocking tendency does not greatly exceed that of the normal Cylinder *B*.

Fig. 15, similar to Figs. 11 to 14, is constructed at a carburetor air-fuel ratio of 11.03 for fuel S-6 which was specially blended to have light and heavy ends of extremely low and high antiknock value respectively. As the rate of fuel vaporization in the manifold is decreased, the inherent antiknock value of the fuel in lean Cylinder *A* is decreased, and the air-fuel ratio of the mixture moves nearer to that for maximum knock, consequently the knocking tendency increases rapidly. Under the same conditions, as the mixture in rich Cylinder *C* moves farther away from the maximum knock point, the inherent antiknock value of the fuel in the cylinder increases. The two effects combine to cause the knocking tendency of the fuel in Cylinder *C* to diminish rapidly. It should be noted

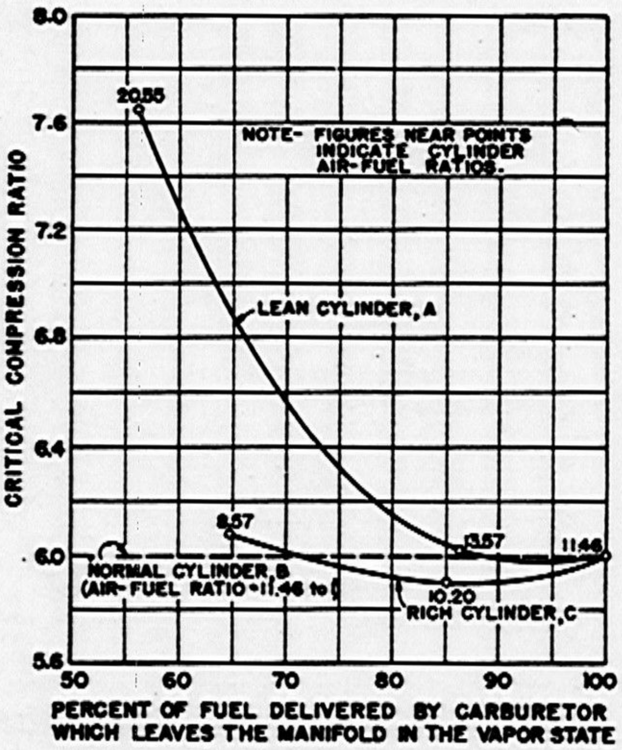

FIG. 13 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

FUEL S-11, SUMMER VOLATILITY, EXTREME CONCENTRATION OF ANTIKNOCK IN LIGHT ENDS.
AIR-FUEL RATIO AT THE CARBURETOR, 11.46

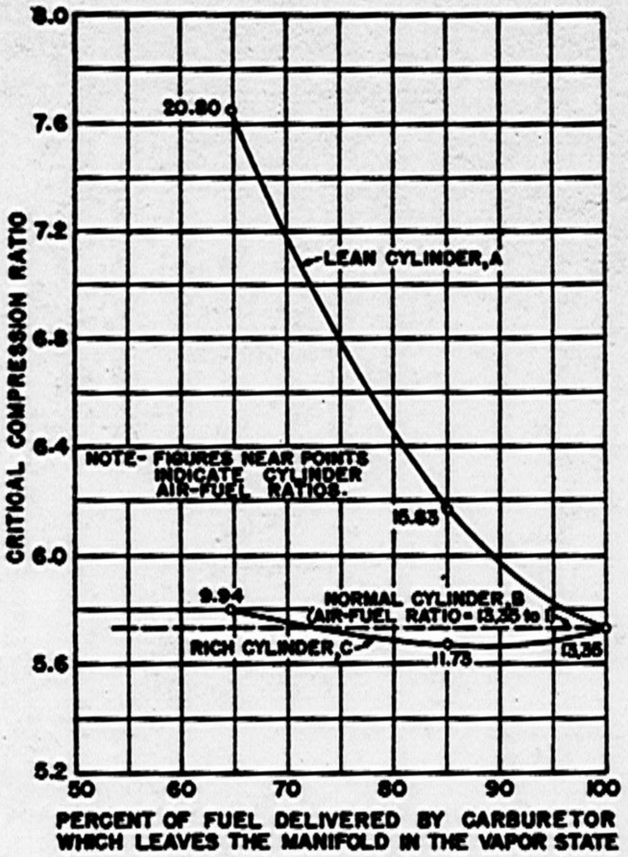

FIG. 14 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

FUEL S-11, SUMMER VOLATILITY, EXTREME CONCENTRATION OF ANTIKNOCK IN LIGHT ENDS. AIR-FUEL RATIO AT THE CARBURETOR, 13.35 (MAXIMUM KNOCK)

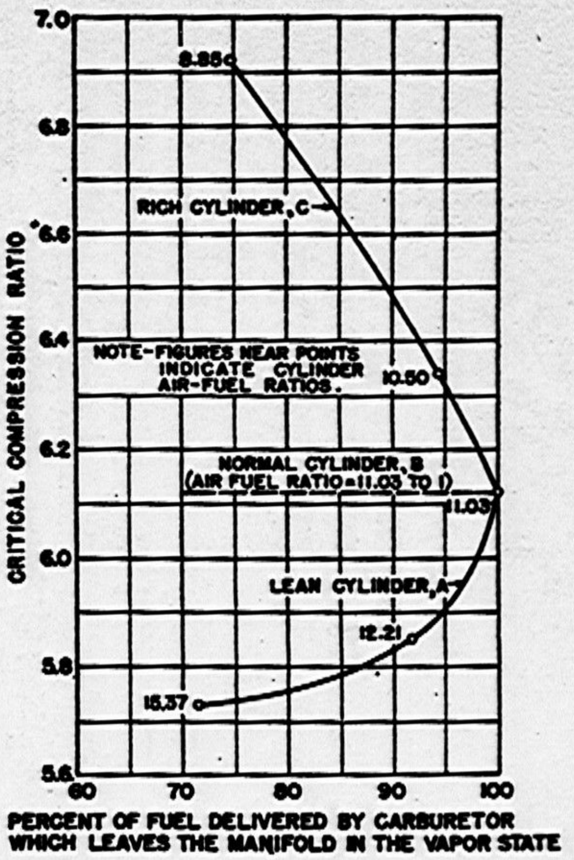

FIG. 15 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

FUEL S-6, WINTER VOLATILITY, EXTREME CONCENTRATION OF ANTIKNOCK IN HEAVY ENDS. AIR-FUEL RATIO AT THE CARBURETOR, 11.03

that at 75 per cent vaporization the difference between the critical compression ratios for Cylinders *A* and *C* is 1.19 ratios, equivalent roughly to 20 octane numbers.

Fig. 16 shows that, when the carburetor air-fuel ratio for fuel S-6 is that for maximum knock, the lean Cylinder *A* has a slightly greater tendency to knock than the normal Cylinder *B* when the rate of vaporization is between 90 and 100 per cent, but that Cylinder *B* knocks most at lower rates of vaporization. The effect of carburetor air-fuel ratio is quite pronounced for this fuel.

Fuels of commercial interest have a distribution of antiknock value along their distillation curves which lies between but does not approach that for fuels S-6 and S-11. A study of the charts for these two fuels, together with those for fuel S.E.G.M. No. 2, should permit a prediction of the behavior of fuels having various degrees of concentration of antiknock value in the light and heavy ends.

Some mention should perhaps be made of a hypothetical fuel, all of whose fractions have equal inherent antiknock value. The knocking tendency of the fuel in any cylinder would depend only on the air-fuel ratio with respect to that for maximum knock. At the carburetor air-fuel ratio for maximum knock, the normal Cylinder *B* would have the greatest knocking tendency, regardless of the degree of vaporization. At a normal carburetor air-fuel ratio, which would be richer than that for maximum knock, the knocking tendency of Cylinder *C* would decrease with a decrease in the rate of vaporization, whereas Cylinder *A* would knock increasingly more as its air-fuel ratio approached that for maximum knock.

Effect of Atmospheric Temperature

In the case of an engine whose choke and mixture-heating valves are not adjusted automatically or manually as the atmospheric temperature changes, the effect of a decrease in atmospheric temperature is two-fold, as follows:

(1) To lean the carburetor mixture.

(2) To decrease the vaporization of the fuel in the carburetor and intake manifold and to widen the differential between the air-fuel ratios of the richest and leanest cylinders.

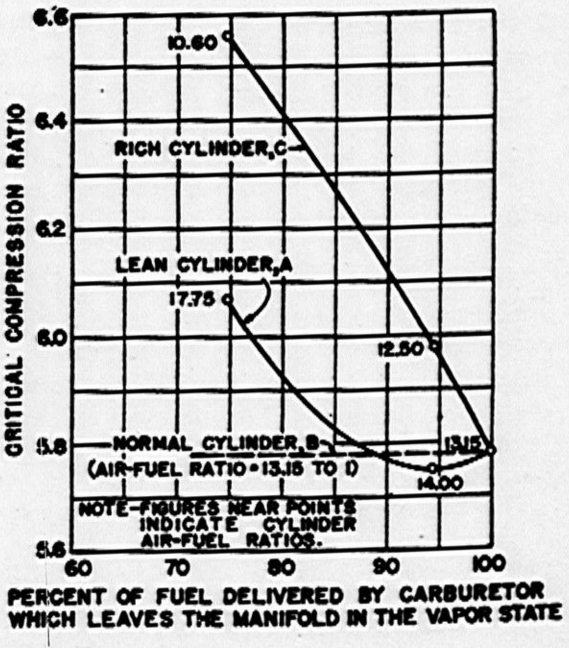

FIG. 16 EFFECT OF UNEQUAL DISTRIBUTION OF THE FUEL IN THE MANIFOLD ON THE CRITICAL COMPRESSION RATIOS OF THE INDIVIDUAL CYLINDERS OF AN ENGINE.

FUEL S-6, WINTER VOLATILITY, EXTREME CONCENTRATION OF ANTIKNOCK IN HEAVY ENDS. AIR-FUEL RATIO AT THE CARBURETOR, 13.15 (MAXIMUM KNOCK)

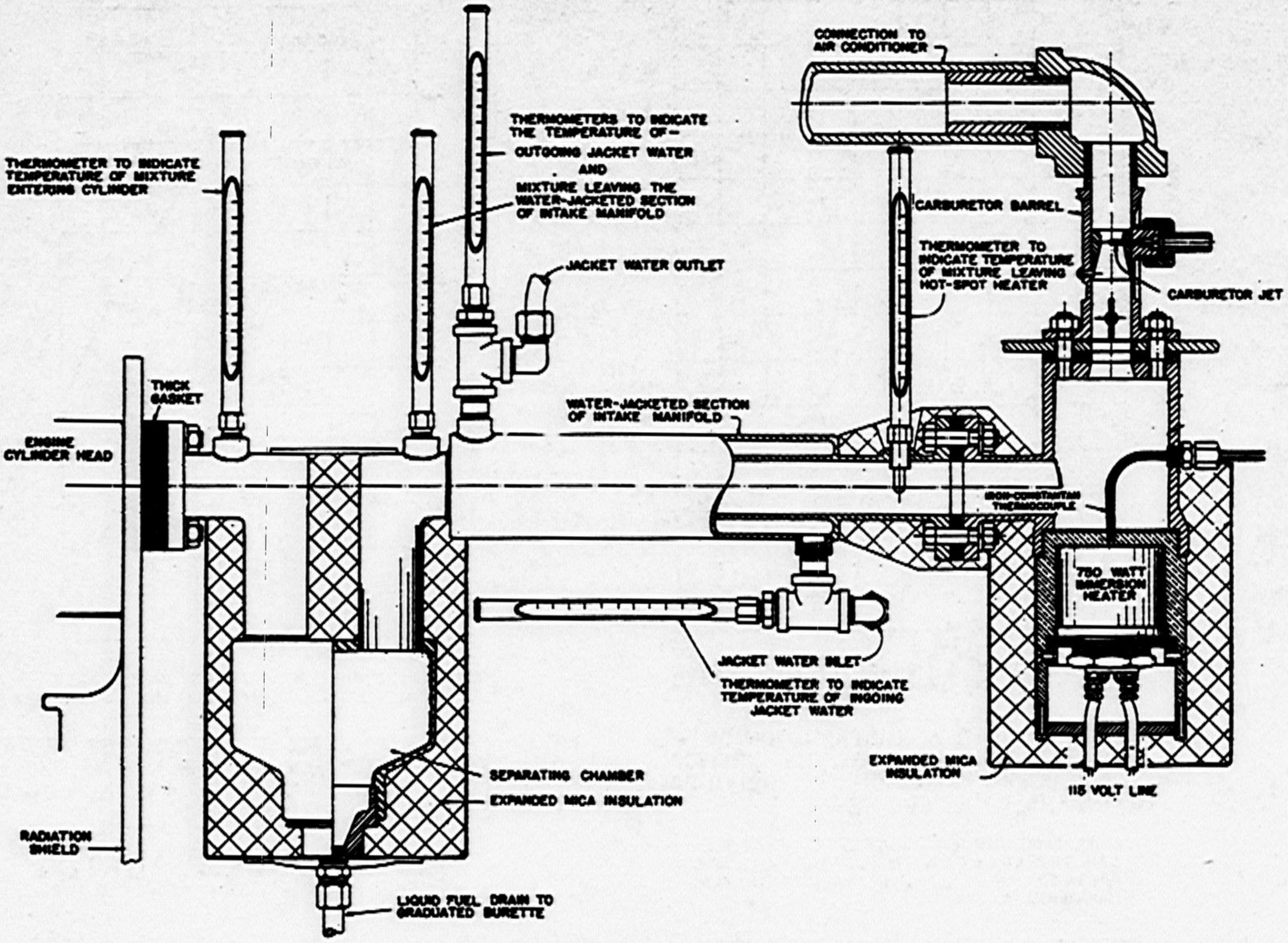
THERMOMETER TO INDICATE TEMPERATURE OF MIXTURE ENTERING CYLINDER
THERMOMETERS TO INDICATE THE TEMPERATURE OF—
OUTGOING JACKET WATER
AND
MIXTURE LEAVING THE WATER-JACKETED SECTION OF INTAKE MANIFOLD
JACKET WATER OUTLET
THICK GASKET
ENGINE CYLINDER HEAD
WATER-JACKETED SECTION OF INTAKE MANIFOLD
CONNECTION TO AIR CONDITIONER
CARBURETOR BARREL
THERMOMETER TO INDICATE TEMPERATURE OF MIXTURE LEAVING HOT-SPOT HEATER
CARBURETOR JET
IRON-CONSTANTAN THERMOCOUPLE
750 WATT IMMERSION HEATER
JACKET WATER INLET
THERMOMETER TO INDICATE TEMPERATURE OF INGOING JACKET WATER
EXPANDED MICA INSULATION
115 VOLT LINE
SEPARATING CHAMBER
EXPANDED MICA INSULATION
RADIATION SHIELD
LIQUID FUEL DRAIN TO GRADUATED BURETTE

FIG. 17 MODIFIED SINGLE CYLINDER ENGINE MANIFOLD AND ASSOCIATED EQUIPMENT FOR THE STUDY OF THE RELATIVE EFFICIENCY OF AIR AND MIXTURE HEATING IN THE VAPORIZATION OF FUEL

Obviously a reduction in atmospheric temperature, in the absence of adjustment of choke or mixture heat-control valve, may result in marked changes in the relative ratings of fuels which bear no relation whatever to the sensitivity of the fuels as determined by the differences between their Research and Motor Method ratings. This point is confirmed by a study of the data from recent road knock tests.

Adjustment of Choke and Mixture Heat Control

Some form of choke, either manual or automatic, always is installed on the car engine. There is usually also a manual or automatic heat-control valve. During the warm-up period in the winter the effect of the choke is to decrease the air-fuel ratio considerably. Under these conditions the degree of vaporization is low, and the relative knocking tendency of the several cylinders of the engine may be predicted by a study of Figs. 11, 13, and 15.

After the warm-up period the choke is usually not an important factor in the knocking problem, but this condition is not true of the mixture heat-control valve, especially if automatically actuated. As applied to one well-known car, the valve bypasses the maximum heat to the intake manifold when the temperature of the air surrounding the manifold is low, but applies progressively less heat as the temperature under the hood rises until, at an air temperature of 75 deg. fahr. near the thermostat, the valve is closed entirely. The temperature of the air at the thermostat is about 60 deg. fahr. higher than that of the air outside the hood. If an outside air temperature of 100 deg. fahr. and a corresponding temperature under the hood of 160 deg. fahr. are required for complete vaporization, a substantial portion of the fuel leaves the manifold in the liquid state when the mixture heat-control valve closes. Vaporization may actually be less when the valve closes than at some lower outside air temperature.

The relative efficiency of heat in vaporizing the fuel, when applied to the carburetor air and to the mixture through a hot-spot, immediately becomes of much interest. Fig. 17 shows diagrammatically a modified single-cylinder engine intake manifold, together with associated equipment, designed for a study of these effects. The manifold has a long horizontal section and a U-bend, the latter being provided with an opening through which liquid fuel flows to a graduated burette. After flowing downward through the vertical carburetor, the fuel and air mixture is forced to turn a corner. Directly under the carburetor is the electrically heated hot-spot against which liquid fuel impinges as it turns the corner.

A section of the manifold is surrounded by a jacket through which water of varying temperature may be circulated for heating of the mixture. Air for the carburetor is supplied by an air-conditioner which is capable of delivering air having a range of temperature and humidity.

Fig. 18 shows for one commercial gasoline the relation between temperature of the mixture entering the cylinder and the degree of fuel vaporization when the heat is supplied alternately to the carburetor air and to the hot-spot. Between the limits of no heat applied to the hot-spot and complete vaporization of the fuel by either method of heating, the mixture at any temperature leaving the manifold contains a greater fraction of the fuel delivered by the carburetor when the heating is done by the hot-spot. For example, at a final mixture temperature of 85 deg. fahr., 88 per cent of the fuel passes into the cylinder with the air when the carburetor air is heated, but the quantity increases to 94 per cent when the heat is applied by the hot-spot. Similarly, complete vaporization of the fuel is accomplished at a lower mixture temperature in the case of hot-spot heating.

Variations in Ignition Timing Between Cylinders

The relative knocking tendency of the cylinders of a multicylinder engine varies with carburetion and fuel distribution, as illustrated in Figs. 11 to 16, provided that spark advance and cylinder cooling are uniform. Because of the longer cylinder jackets and better direction of flow of cooling water in recently built car engines, it is believed that variations in knocking among the cylinders due to differences in cooling have, in general, been minimized. Variations in spark advance appear to create a greater problem. For example, the leanest cylinder may have the greatest knocking tendency because of partial fuel vaporization and imperfect distribution, as illustrated in Fig. 11. If, however, the spark advance in this cylinder during knocking is retarded relative to that in the other cylinders, the most pronounced knock may occur in some other cylinder. In any study of the effects of fuel distribution it is desirable, therefore, to be able to measure the ignition advance in each cylinder simultaneously.

Fig. 19 shows a recently developed instrument for this purpose. A drum weighing 20 lb. and designed to hold a strip of 35-mm. cinema film on its outside surface is driven at a constant speed of 75 r.p.m. by a 6-volt electric motor. A neon tube is enclosed in a small light-tight box which is quite close to the drum. A narrow slot in the box, parallel to the axis of the film drum, permits the flash of the neon tube to appear as a line on the film, perpendicular to its length. The tube lights as a result of the voltage induced by the breaking of the primary circuit of the ignition system and by the motion of two flywheel studs, 180 deg. apart, past a magnetic pickup mounted on the flywheel housing. The pickup is similar in design to that employed by the Chrysler Corp. in its instrument for the measurement of mean spark advance in the cylinders. The induced voltage is amplified by the triode section of a duplex diode-triode vacuum tube (Type 6R-7) and the amplified output is coupled to the grid of a power tube (Type 6A-3). The neon tube is connected in parallel with the load resistance of the power tube. It is lighted when the voltage across the load resistance of the power tube rises to approximately 90 volts and is extinguished when the voltage applied to the grid of the power tube decreases due to termination of the signal impulse.

The oscillatory impulse from the primary of the ignition coil due to the opening of the breaker points is rectified and amplified in the Type 6R-7 tube and applied to the grid of the power tube, producing a flash of the neon tube and a mark on the film for the firing of each cylinder. A portion of the high-voltage impulse to No. 1 spark-plug is amplified and applied to the grid of the power tube, resulting in a flash of longer duration which identifies the film mark for No. 1 cylinder. One revolution of the film drum covers a number of complete engine cycles. A ratchet-type relay limits the exposure to one revolution of the film drum.

Ignition timing of the individual cylinders is read from a curve of angular displacement of the crankshaft versus time, obtained from the flywheel reference points.

Knocking Tendency of Individual Cylinders

The knocking tendency of the individual cylinders of a car engine may be determined by the aid of the auxiliary breaker used with the gas sampling valve as shown in Fig. 2. When the knocking of No. 1 cylinder is to be measured, the regular distributor is retarded and the auxiliary breaker is set to produce a timing in No. 1 cylinder which is the same as that given by the regular distributor when adjusted properly. After the knocking tendency of No. 1 cylinder has been measured, the auxiliary breaker is used to produce the correct ignition timing in Cylinder No. 2, and so on for the remainder of the cylinders. Since the auxiliary breaker is mounted on the same shaft as the regular breaker, it duplicates the advance characteristics of the regular breaker.

Reference Fuels

The scale of ordinates for Figs. 11 to 16 is critical compression ratio. For some purposes it may be desirable to substitute an octane-number scale, but many such scales are possible, depending on the choice of reference fuels and the conditions under which they are tested. For example, either the primary or secondary reference fuels might be used. They might be run always at the maximum knock air-fuel ratio or at the same carburetor setting as for the fuel under test. A blend of the secondary reference fuels equal to a given blend of the primary reference fuels at the maximum knock air-fuel ratio may be far from equal at another air-fuel ratio.

The reference fuels may be run under conditions of complete vaporization or under the manifold conditions employed for the fuels under test. In the usual tests of commercial

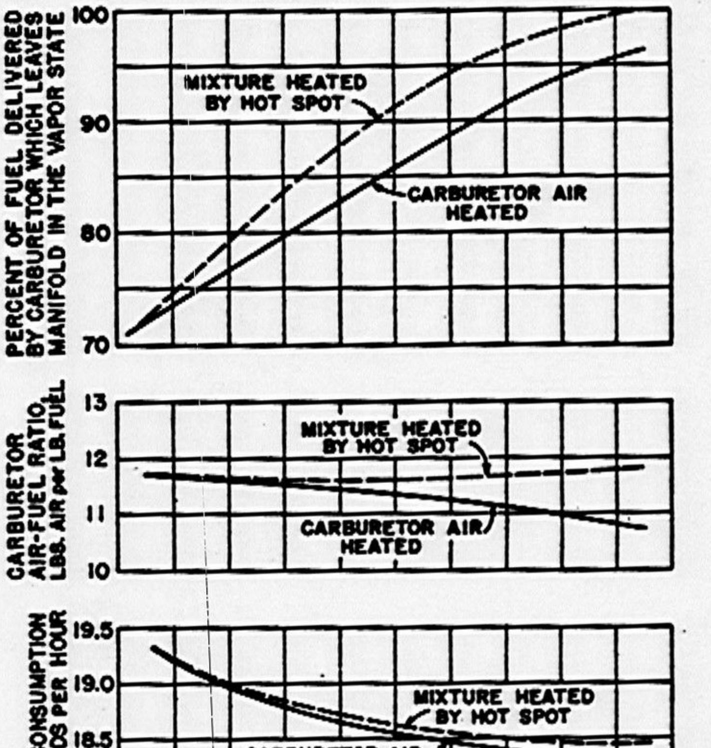

FIG. 18. EFFECT OF HOT SPOT MIXTURE HEATING AND CARBURETOR AIR HEATING ON THE VAPORIZATION OF FUEL IN THE MANIFOLD.

SERIES 30B ENGINE WITH SPECIAL MANIFOLD AND EQUIPMENT OF FIG. 17.
FIXED CARBURETOR SETTING
ENGINE SPEED, 900 R.P.M.

gasolines, only a small fraction of the primary reference fuels fails to vaporize, even when the rate of vaporization of the test fuels is low. Under the same conditions, a much larger fraction of the secondary reference fuel blends remains in the liquid state and the unvaporized portion is composed largely of fuel A-5. The equivalent octane numbers of single points on the curves of Figs. 11 to 16 may vary as much as 15 octane numbers, depending on the choice and method of testing of the reference fuels. Curves of octane number versus per cent of vaporization may have shapes entirely different from the corresponding curves of critical compression ratio versus per cent of vaporization.

There is every reason to believe that the complication introduced into the interpretation of laboratory octane numbers obtained under conditions of partial fuel vaporization enters also into the determination of road octane numbers. Regular-grade gasolines require the use of the C-10 and A-5 reference fuels. As the per cent of the A-5 component in the blend is increased, the fraction of the blend which is vaporized in the manifold should decrease, causing the leanest cylinder to become leaner and the richest cylinder richer. The cylinder which knocks most on the test fuel may not knock most on the reference-fuel blend, and the cylinder which knocks most on a blend containing a high per cent of C-10 may not knock most when the per cent of C-10 is reduced.

If the volatility of the two secondary reference fuels were interchanged, the knocking tendency of a given percentage blend under any condition of imperfect distribution obviously might be markedly different. An increase in the concentration of A-5 in the blend would then *increase* the vaporization in the manifold with a resultant *decrease* in the inherent antiknock value of the vaporized portion. Such effects introduced into the road knock-testing procedure by the difference in volatility of the reference fuels appear in the road ratings of the test fuels as apparent appreciation or depreciation of these fuels with respect to the laboratory ratings.

Once the observation is made that road octane numbers determined under conditions of imperfect manifold vaporization and fuel distribution are influenced by the choice of secondary reference fuels, a consideration of desirable characteristics of secondary reference fuels assumes real importance. In the light of the previous discussion it appears that, from the standpoint of minimizing the complications introduced into road knock testing by the secondary reference fuels, such fuels ideally should have the following characteristics:

(1) The distillation curve of the fuel of high antiknock value should match exactly that of the fuel of low antiknock value in order to provide constant vaporization in the manifold as the proportions of the two fuels in the blend are changed.

(2) The volatility of the reference fuels should approximate that of the average commercial gasoline, thereby ensuring, in so far as possible, the same cylinder air-fuel ratios for the test and reference fuels.

(3) All fractions of each fuel should have equal antiknock value in order to prevent a change in the inherent antiknock value of the vaporized portion of the blend in the manifold as the degree of vaporization is changed. Inasmuch as commercial gasolines have light ends whose antiknock value is higher or lower than or equal to that of the heavy ends, the distribution of antiknock value in the reference-fuel blends would then approximate that of the average of fuels met in service.

These specifications constitute a large order which will probably never be filled. It should, however, be possible to produce secondary reference fuels which, for use under conditions of partial vaporization, are an improvement over the present A-5 and C-10 fuels. The latter fuels are entirely satisfactory for use in the laboratory in conjunction with the standard C.F.R. engine manifold and were almost equally suitable for road-test purposes until the fuel-distribution prob-

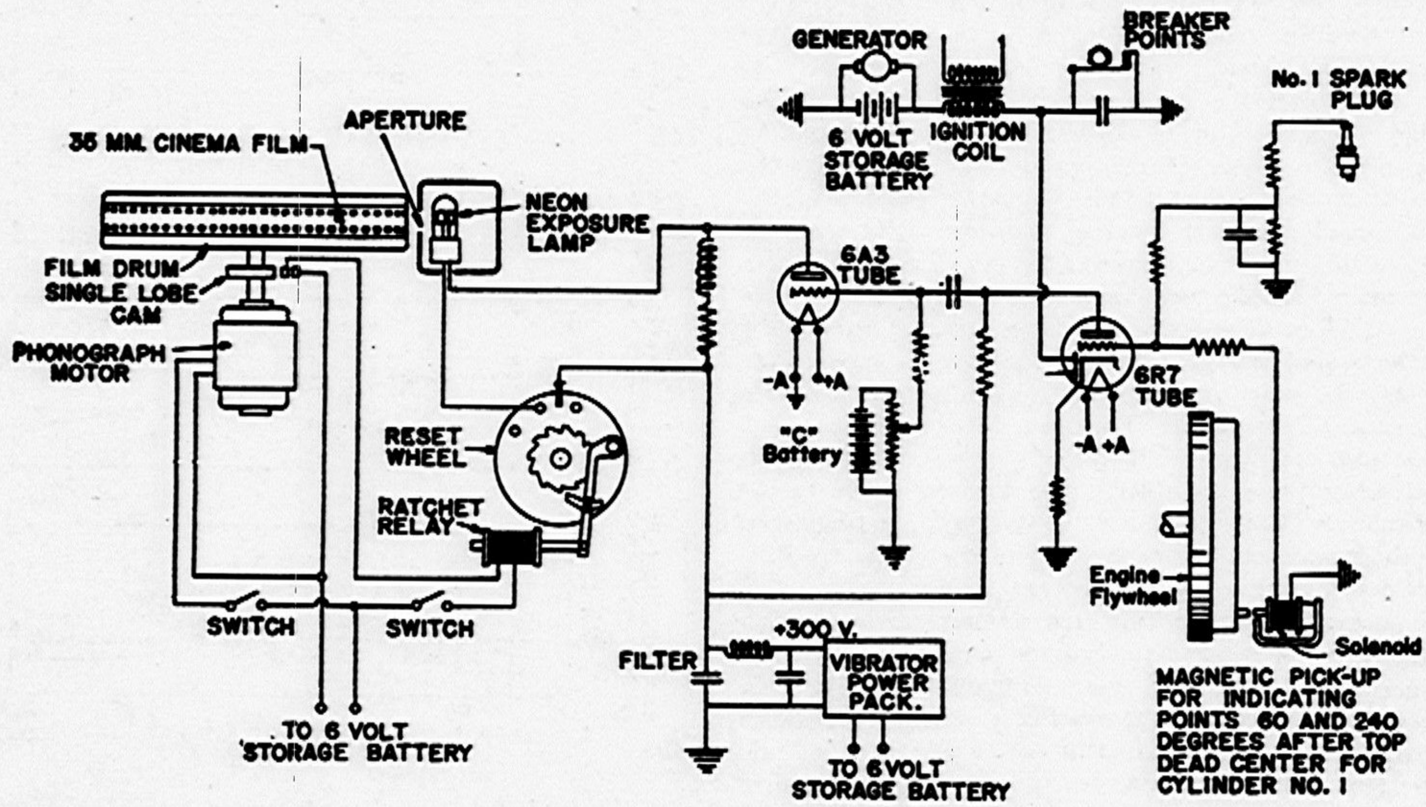

FIG. 19 DIAGRAMMATIC SKETCH OF AN INSTRUMENT FOR RECORDING ON CINEMA FILM THE IGNITION TIMING IN THE INDIVIDUAL CYLINDERS OF A MULTICYLINDER ENGINE.

lem attained its present importance. It should be noted that the primary reference fuels substantially fulfill the first and third of the above-described specifications.

Correlation of Road and Laboratory Ratings

The correlation of laboratory with road knock ratings creates two distinct and basically different problems, namely:

(1) Reproducing on the laboratory engine the conditions of operation of a single car engine in a single test.

(2) Determining a laboratory procedure which gives ratings that agree in so far as possible with the averages of ratings obtained in a group of cars under one or more conditions of test.

The first problem is by far the simpler. If every variable of engine operation such as mixture and cylinder temperatures, spark advance, speed, fuel composition, and mixture ratio, applicable to the knocking cylinders of the multicylinder engine is duplicated on the laboratory engine, the ratings of all types of fuels should be duplicated provided, of course, that the same reference fuels are used in both cases.

If partial fuel vaporization in the intake manifold of the car engine gives rise to differential distribution of unvaporized liquid fuel which, in turn, causes a cylinder richer or leaner than the normal cylinder to have the greatest knocking tendency, the knock ratings in the car engine of a wide variety of fuels can not be duplicated on the laboratory engine unless provision is made for the simulation of the mixture distribution of the multicylinder engine by some means such as that described herein. A fortuitous compensation for the effects of mixture distribution may be made for a limited number of fuels by changes in other engine variables, but such an arrangement is almost certain to fail when applied to a large group of fuels.

A modification of the conventional laboratory knock-testing procedure, by the substitution of a manifold designed to simulate the action of the intake manifold of the car under consideration should go a long way toward explaining the anomalous behavior of fuels observed from time to time in a single car.

Before an attempt is made to duplicate on the laboratory engine the ratings obtained on a given car, it is believed advisable to construct a chart similar to Fig. 20 in which air-fuel ratio, spark advance, and cylinder knock intensity are plotted against cylinder number. Such a chart not only indicates the cylinder of greatest knocking tendency and its air-fuel ratio, but furnishes the data required for the computation of average air-fuel ratio and the degree of vaporization. The information on ignition timing may serve to explain the greatest knocking tendency of one cylinder when, under conditions of uniform ignition timing, the laboratory engine indicates that a cylinder having a different mixture condition should knock most.

The second problem – that of finding a set of operating conditions for the laboratory engine which gives ratings that agree with the average of ratings in a group of cars – is quite different. The average road ratings reflect the results of the simultaneous operation of a host of variables combined differently in each car. Considering alone those variables which arise through variations in carburetion and fuel distribution in the engine, it is obvious that a fuel might be rated in one car under conditions of substantially perfect distribution and at an air-fuel ratio near that for maximum knock whereas, in another car, distribution might be equally good but the air-fuel ratio in all cylinders might be considerably richer than that for maximum knock. In a third car with imperfect fuel distribution the rating might be determined by the behavior of the portion of the fuel leaving the manifold in the vapor state and burned at the air-fuel ratio for maximum knock. The list of possibilities is far from exhausted when it becomes apparent that no single set of carburetor and manifold conditions for the laboratory engine theoretically should give ratings equal to the average of ratings obtained on several cars having a wide variation in carburetion and manifold conditions.

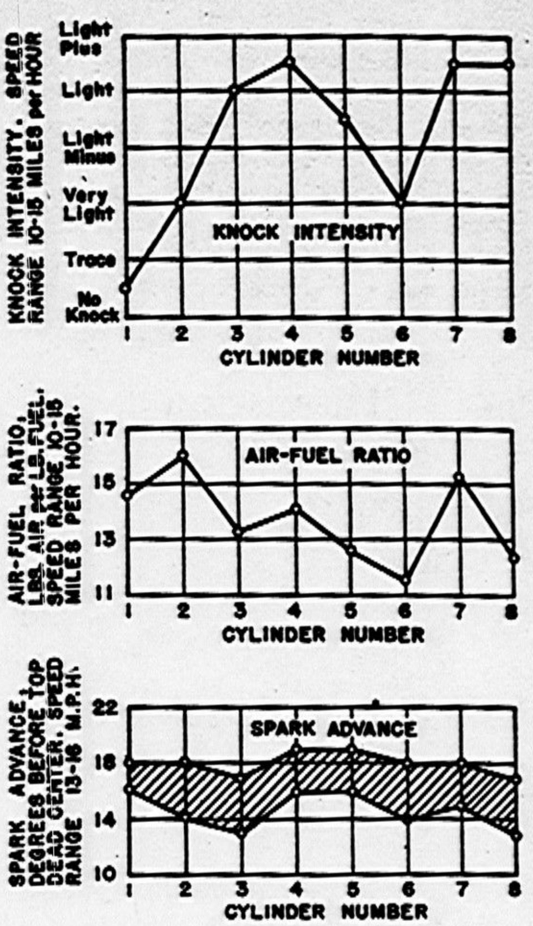

FIG. 20. VARIATION OF AIR-FUEL RATIO, SPARK ADVANCE, AND KNOCK INTENSITY AMONG THE CYLINDERS OF AN AUTOMOBILE ENGINE IN THE SPEED RANGE OF MAXIMUM KNOCKING.

Notwithstanding this situation, definite and consistent effects of non-uniform fuel distribution are now apparent in any fuel knock-rating data obtained on a group of cars. One of the most impressive demonstrations of the importance of this variable was furnished by a series of road tests in which the 12 test fuels were specially blended to have an extreme concentration of antiknock value in different portions of the distillation range. Fuels S-6 and S-11, previously mentioned, were two of the test fuels.

After the effects of imperfect fuel distribution within the engine have been found to exist, the problem of most immediate interest is the development of some means for the correction of variations in the road ratings of fuels, rated as equal by present laboratory methods, due to this variable. The British have for some time used a correction factor for benzol blends. For the first 30 per cent of the total aromatics in any gasoline, 1 octane number is added to the conventional laboratory rating and, for each additional 15 per cent of aromatics, another octane number is added. A correction factor applicable to all types of fuels and capable of direct determination on the engine would be desirable.

It is to be hoped that automotive engineers will be able to find a not-too-expensive means for minimizing variations in fuel distribution at low engine speeds. In the meantime the oil refiner will undoubtedly make an effort to correct, by some change in the laboratory knock-testing procedure, present inequalities in the road performance of fuels of equal octane number arising because of the mixture-distribution problem.

The addition of concentrated knock suppressors to fuels may change the distribution of antiknock value through the fuel fractions. The choice of antiknocks to meet the wide range of conditions resulting from variations in carburetion and manifolding is a complex problem in itself. It is hoped that this subject may be discussed in a later paper.

Discussion

Stresses Need for Further Study

– L. E. Hebl

Shell Petroleum Corp.

We must recognize that, in our study of detonation, none of us has followed the scientific method more closely than has Mr. Bartholomew, whose evaluation and classification of the variables involved has been relentless. He was probably the first to begin classifying primary and secondary variables when in 1929 he said: "In fact the variation in ratings with changes in speed, spark advance, compression ratio, throttle opening, and so on can, in almost every case, be traced to a difference in temperature conditions."[*] Now again he pioneers in demonstrating that not only those variables influencing the manner in which the mixture burns within the cylinder are significant, but also account must be taken of the variables that affect the composition of the mixture that enters the cylinder.

Mr. Bartholomew's paper comes at a very opportune time when the automobile and oil industries are disturbed by the realization that the present test method does not rate fuels in the correct order of their performance in multicylinder engines. Although the Cooperative Committee has already carried out some tests which could probably be used as a basis for modifying the test method, it would be very unwise to proceed with the adoption of a modified test until Mr. Bartholomew has had time to examine the manifold phenomena more thoroughly and to learn whether a practical correction for these phenomena can be applied to the laboratory test.

Briefly, it now appears that the manifold phenomena tend to lower the ratings in multicylinder engines and, in order to obtain agreement, the single-cylinder laboratory engine has been operated at excessively high temperatures so that ratings in it also are reduced to match the average road values. However, the high-temperature affects cracked gasolines mainly, whose ratings are reduced too much in the laboratory, whereas ratings on straight-run gasolines, which are insensitive to temperature, are not reduced enough. The resulting deviations are thus both positive and negative so the average appears to be correct; however, if the errors on individual fuels are to be corrected, it is obviously necessary to reduce the temperature of the laboratory engine to a value in line with multicylinder engines and to correct for the lower multicylinder ratings by correcting the same factors that cause those ratings to be low. Mr. Bartholomew has demonstrated a possible source of those errors and it seems logical that a full understanding of them is necessary before further action should be taken.

Caution Urged in Interpreting Results

– A. J. Blackwood

Standard Oil Development Co.

We wish to throw out a word of caution regarding one phase of the discussion and that is "to watch every step when interpreting volatility and octane number of the different fuel fractions as related to road behavior."

It would appear strange that, at this relatively late date, fuel volatility and octane-number distribution should assume any great importance not hitherto apparent, particularly in view of the reasonably good correlations existing between laboratory fuel ratings and road performance. Fuels which in the past have not given the expected road performance as predicted from laboratory ratings have not been, as a rule, fuels having unusual volatility or unusual octane-number distribution throughout the boiling range.

Some work conducted at our laboratories along somewhat similar lines to the multicylinder work described in the paper has indicated that the qualitative distribution factor may not be anywhere near as important as we had previously anticipated. For example, individual exhaust-gas analyses were made on an eight-cylinder engine when burning blends of reference fuels C-10 and A-4. For a range covering the 50-50 blend up through straight C-10 fuel, the air-fuel ratio of any given individual cylinder remained constant although the various cylinders differed widely in themselves. In other words, if say No. 1 cylinder received a 14:1 mixture ratio and No. 7, a 11:1 mixture ratio of the 50-50 blend, the received the same mixture ratios 14:1 and 11:1 with the straight C-10. Keep in mind that these two reference fuels differ widely in volatility and antiknock value.

Now, coincident with the determination of mixture ratio, values for the apparent octane-number requirement of the individual cylinders were determined. These data on the 8 individual cylinders, when plotted against the corresponding air-fuel ratio, gave a smooth curve very closely similar to what would have been obtained if the octane-number requirement of a single-cylinder engine were determined for different mixture ratios. These data were all obtained at full throttle at the speed of maximum knock.

This result would indicate that there had been no appreciable deviations due to qualitative distribution but rather that, if the vaporized fuel is distributed unevenly to the various cylinders, the liquid particles are distributed over the same general pattern.

Therefore, we urge that haste be made slowly when attempting to tie fuel volatility in with road antiknock behavior.

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[*] See *Oil and Gas Journal*, Oct. 17, 1929, p. 32: "Antiknock Testing and Performance," by Earl Bartholomew.

Feb. 23, 1926. 1,573,846

T. MIDGLEY, JR

METHOD AND MEANS FOR USING MOTOR FUELS

Filed April 15, 1922

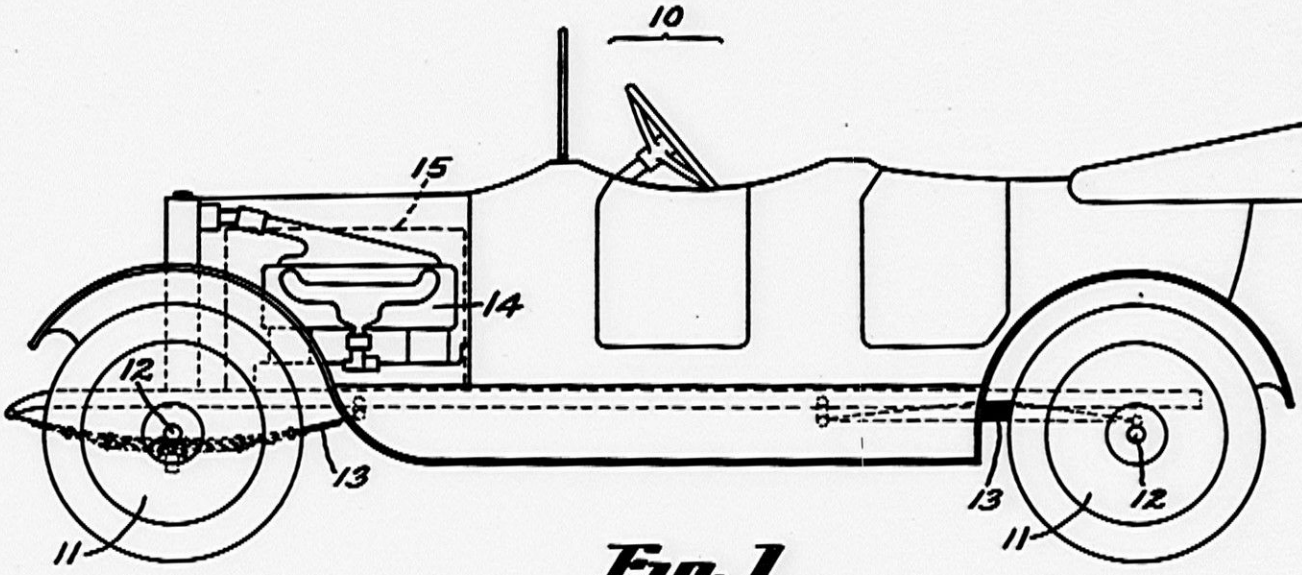

Fig. 1

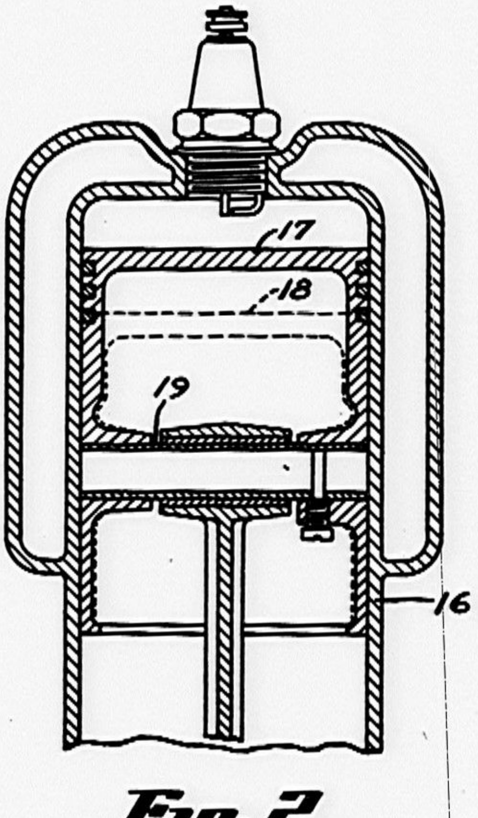

Fig. 2

Witnesses
Irvin A. Greenwald
Lloyd M. Keighley

Inventor
Thomas Midgley Jr.

By Francis D. Hardesty
Attorney

Patented Feb. 23, 1926. **1,573,846**

UNITED STATES PATENT OFFICE.

THOMAS MIDGLEY, JR., OF DAYTON, OHIO, ASSIGNOR, BY MESNE ASSIGNMENTS, TO GENERAL MOTORS CORPORATION, OF DETROIT, MICHIGAN, A CORPORATION OF DELAWARE.

METHOD AND MEANS FOR USING MOTOR FUELS.

Application filed April 15, 1922. Serial No. 553,270.

To all whom it may concern:

Be it known that I, THOMAS MIDGLEY, Jr., a citizen of the United States of America, residing at Dayton, county of Montgomery, and State of Ohio, have invented certain new and useful Improvements in Methods and Means for Using Motor Fuels, of which the following is a full, clear, and exact description.

This invention relates to fuels, such, for example as kerosene and gasoline, employed in the operation of internal-combustion engines and to the art of burning the fuels in an engine. The present tendency is to produce lower grades of gasoline in order to obtain a sufficient output for the increasing demand for motor fuels and to reduce the compressions of the engines so that these lower grades of fuel may be used without knocking. As the lowering of engine compression reduces the efficiency of the engine, a still greater output of fuel is required to meet the increase in fuel required to operate larger and less efficient engines. The principal objects of the present invention are to overcome these difficulties and to provide a means for using either low or high grades of motor fuel more efficiently and so reduce the quantity of fuel used.

The present application is a continuation in part of my application Serial No. 464,-985, filed April 27, 1921.

In the accompanying drawings:

Fig. 1 is a side view of an automobile having an engine embodying the present invention and showing the comparative sizes of engines employed when using high and low engine compressions; and

Fig. 2 is a longitudinal sectional view of an engine cylinder and shows the comparative sizes of combustion chambers employed when using fuels having high and low critical compression pressures.

Kerosene, gasoline and the heavier hydrocarbons have the characteristic that, when a combustible gaseous mixture containing one of these fuels and air is burned in an internal-combustion engine while subjected to a relatively high pressure, a fuel knock is produced, the engine heats rapidly, the efficiency of the engine is reduced and, if the initial pressure is very high, engine parts may be injured. The highest pressure at which a mixture may be burned in a cylinder without producing a fuel knock varies with the different fuels and, to some extent, with the temperature, position of spark plugs and other conditions within the engine. This pressure I term the "critical compression pressure" of the fuel.

The average critical compression pressure of kerosene is about 50 pounds, of the proper grades of gasoline about 75 pounds and of the better grades of gasoline about 125 pounds. The latter grade of gasoline is produced in limited quantities and is not available universally to the consumer. The commoner grades of fuel, such as kerosene and gasoline, having critical compression pressures below 75 pounds are used generally, and in internal-combustion engines for house lighting systems, trucks, tractors, and automobiles are designed to operate on these kinds of fuel.

I have found that the critical compression pressure of a fuel of the type mentioned above is increased by incorporating therewith any one of a large number of compounds containing metallic elements, i. e., compounded metallic elements.

By way of example, I may use a fuel consisting by volume of ¼ of one percent of tetra ethyl lead and 99¾ percent of gasoline having a normal "critical compression pressure" of about 75 pounds. The tetra ethyl lead dissolves in the gasoline forming a fuel having a "critical compression pressure" of about 160 pounds. The presence of the lead compound changes the gasoline from a low compression fuel to a higher compression fuel, i. e., increases its "critical compression pressure." This gasoline may be used in an engine having a compression pressure of about 160 pounds, with a smaller fuel consumption for obtaining a given amount of work than is required to operate an engine having a compression pressure of 75 pounds on the untreated gasoline. Experiments with automobiles have shown that if the engine compression is increased to 160 pounds, a treated fuel being used to run the engine and the gear ratio between the engine and the rear axle is reduced, the mileage obtained from a gallon of gasoline is substantially twice that obtained when running the engine at 75 pounds compression on the untreated or low compression fuel.

In the drawings, 10 is an automobile having wheels 11, axles 12, springs 13, and an engine 14 whose compression pressure is about 160 pounds. This engine may be operated on the mixture of gasoline and tetra ethyl lead just described. At 15 is shown diagrammatically the comparative size of an engine operating on an engine compression of about 75 pounds and capable of delivering the same horse power as engine 14. It is apparent that by employing engine 14 instead of engine 15, the size and weight of the wheels, axles and other parts of the automobile may be materially lightened, and that by lightening the weight of the machine as a whole a still smaller engine may be employed to propel the vehicle.

Fig. 2 illustrates a method of changing the compression of an engine to obtain these results. In this figure, 16 is an engine cylinder and 17 the engine piston in its top dead center position in which the gaseous mixture is compressed to about 160 pounds. The dotted line 18 indicates the top dead center position of a piston which compresses the gases to about 75 pounds. A change from a low compression engine to a high compression engine may be made by substituting a piston whose length from the wrist pin 19 to the top of the piston is greater than in pistons in common use. This change in the pistons increases the horse power of the engine. Experience has shown that operation of an engine of the type illustrated in solid lines in Fig. 2, on a low compression fuel such as common gasoline, gives excessive heating, violent knocking, and under continued operation cracks the piston.

For some commercial purposes it will probably be sufficient to use but one part of tetra ethyl lead in 2000 parts of gasoline. This mixture may be used in a truck motor to avoid the knock generally produced when climbing hills at low speed with open throttle. Relatively small proportional quantities of the lead compound are required to increase the critical compression pressure of "aviation" gasoline from 125 pounds to 160 pounds or more.

The fuel mixture or combination may also be varied by using other fuels as the fuel base or major constituent of the high compression fuel and using other compounded metallic elements to increase critical compression pressure. The fuels used include gasoline and other hydrocarbon fuels containing hydrocarbons which may be obtained from an asphalt, naphthene paraffine base crude or mixtures thereof.

Other lead compounds which have been used successfully are the phenyl compounds and other alkyl compounds besides the tetra ethyl lead. These include the ethyl, isopropyl and methyl compounds. Methods of manufacturing lead compounds are described on page 133 et. seq., vol. 111 of Watts' Dictionary of Chemistry as revised by Morley and Muir and published by Longmans, Green and Co., London, England.

Other metallic compounds may be used to suppress a knock in a motor. These include compounds of selenium, tellurium, tin, arsenic and antimony. The phenyl and alkyl compounds of these metals have been successfully used where the stable compounds are obtainable. The alkyl metal compounds usually have a greater knock suppressing value than the corresponding phenyl compounds.

Dissolving a knock suppressing substance in a hydrocarbon fuel has been found the most satisfactory method of changing the critical compression pressure of the fuel, and for this reason the organic compounds have been employed to a greater extent than other compounds in suppressing knocks, however, I do not limit my invention to organic compounds nor to the oil soluble compounds.

The value of metallic compounds in changing the critical compression pressure depends upon the metal used, the composition of the compound, its stability and volatility. Up to the point of complete vaporization the anti-knock value depends in part on the percentage of the compound vaporized under engine condtions. Where the nature of the compound changes before ignition of the fuel, combustion of the fuel takes place in the presence of the new compound and the anti-knock value depends on the nature of the new compound instead of the compound added to the fuel.

If a radical is attached to the metal in the compound in such a manner that the resulting compound is oil soluble and volatile, a high anti-knock value for that metal is obtained. As the compound departs from these conditions either in the kind of radical employed or the number of radicals of a certain kind in the compound, the anti-knock value will generally be reduced. Examples of compounds in which the anti-knock value, as based on these conditions, becomes negligible are lead oleate and lead acetate. If, in the formula for tetra-ethyl lead, an OH radical is substituted for an ethyl radical, the resulting compound (tri ethyl lead hydroxide) is less volatile and less soluble in the fuel than the tetra ethyl lead. The tri-ethyl lead hydroxide is almost as powerful a knock-suppresser as the tetra ethyl lead.

The value of a compounded metallic element as a knock suppresser appears also to be a period function of the atomic number and, in general, with similar elements increases with increasing atomic weight. To illustrate this function, I take the ethyl compounds of the metals in the right hand column of the 4th, 5th, and 6th groups, wherever stable ethyl compounds are obtainable. As the ethyl compound of antimony is unstable in the air at room temperature, I employ the phenyl compound of antimony. These compounds increase in knock suppressing value in group 4 in the order tin, lead; in group 5 in the order arsenic, antimony; and in group 6 in the order selenium, tellurium. Taking group 4 as an example, 1% by volume of the ethyl compounds of the metals in gasoline or kerosene give the following increases in critical compression pressure: tin, 15 pounds; lead at the rate of 340 pounds when employed in relatively small quantities.

While I have mentioned more particularly the use of phenyl and alkyl compounds of certain metals for increasing the critical compression pressure of kerosene and gasoline, my invention is not limited wholly to the use of these compounds, nor to compounds of these metals, and the compounded metallic elements may be employed with the heavier hydrocarbon or other fuels.

Further, while the specific examples herein given constitute preferred forms of embodiment of the invention, it is to be understood that other forms might be adopted, without departing from the scope of the claims which follow.

What I claim is as follows:

1. A fuel for internal combustion engines comprising a low compression motor fuel, and a volatile compound of a metal adapted to increase the critical compression pressure of the motor fuel.

2. A fuel for internal combustion engines comprising a low compression motor fuel, and an oil soluble volatile compound of a metal adapted to increase the critical compression pressure of the motor fuel.

3. A fuel for internal combustion engines comprising a low compression motor fuel, and a relatively small amount of compound consisting of a metal bound to a hydrocarbon radical.

4. A fuel for internal combustion engines comprising a low compression motor fuel, and a relatively small amount of a volatile compound consisting of a metal bound to a hydrocarbon radical.

5. A fuel for internal combustion engines comprising a low compression motor fuel, and a relatively small amount of oil soluble compound consisting of a metal bound to a hydrocarbon radical.

6. A fuel for internal combustion engines comprising a low compression motor fuel, and a relatively small amount of a volatile and oil soluble compound consisting of a metal bound to a hydrocarbon radical.

7. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of an alkyl compound of a metal containing more than two alkyl groups.

8. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of an oil soluble compound of a metal containing more than two alkyl groups.

9. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of a volatile compound of a metal containing more than two alkyl groups.

10. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of a volatile and oil soluble compound of a metal containing more than two alkyl groups.

11. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of an alkyl compound of a metal containing more than three alkyl groups.

12. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of an oil soluble compound of a metal containing more than three alkyl groups.

13. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of a volatile compound of a metal containing more than three alkyl groups.

14. A fuel for internal combustion engines comprising a hydrocarbon motor fuel containing a relatively small proportion of a volatile and oil soluble compound of a metal containing more than three alkyl groups.

15. A fuel for internal combustion engines comprising kerosene or gasoline; and a volatile alkyl compound of a metallic element homogeneously incorporated therein.

16. A fuel for internal combustion engines comprising a low compression motor fuel; and a volatile ethyl compound of a metallic element homogeneously incorporated therein and adapted to be vaporized in an internal-combustion engine before combustion of the fuel therein.

17. A fuel for internal combustion engines comprising a low compression motor fuel; and an alkyl compound of a metal in the right hand column of a group between the third and seventh groups of Mendeleef's table.

18. A fuel for internal combustion engines comprising a low compression motor fuel; and a volatile lead compound homogeneously incorporated therein.

19. A fuel for internal combustion engines comprising a low compression motor fuel; and an alkyl compound of lead.

20. A fuel for internal combustion engines comprising kerosene or gasoline; and an ethyl compound of lead.

21. A fuel for internal combustion engines comprising gasoline; and tetraethyl lead.

In testimony whereof I hereto affix my signature.

THOMAS MIDGLEY, Jr.

Certificate of Correction.

It is hereby certified that in Letters Patent No. 1,573,846, granted February 23, 1926, upon the application of Thomas Midgley, Jr., of Dayton, Ohio, for an improvement in "Methods and Means for Using Motor Fuels," an error appears in the printed specification requiring correction as follows: Page 1, line 62, for the word "proper" read *poorer;* and that the said Letters Patent should be read with this correction therein that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 22d day of June, A. D. 1926.

[SEAL.]

M. J. MOORE,
Acting Commissioner of Patents.

Dec. 5, 1933. F. W. SULLIVAN, JR., ET AL 1,938,547

LEADIZED GASOLINE

Filed March 21, 1932

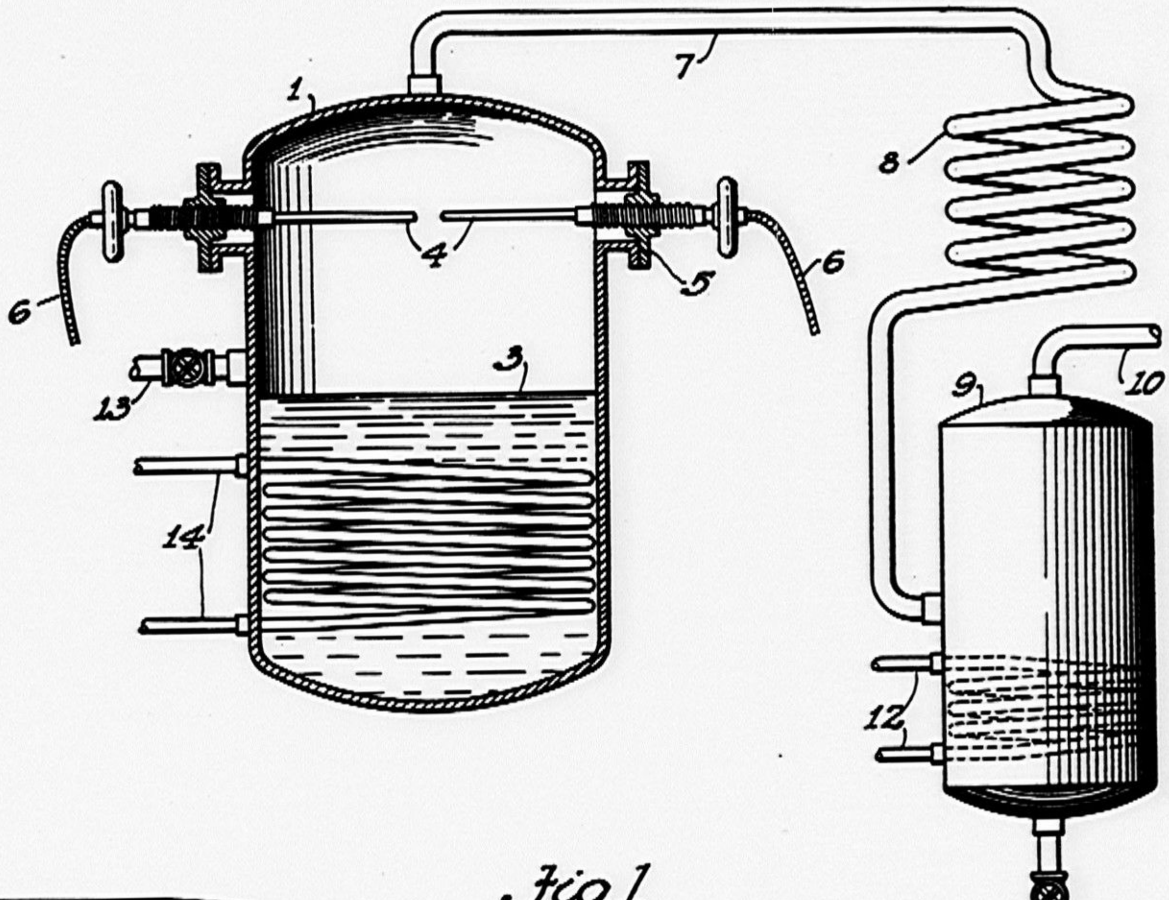

Fig. 1

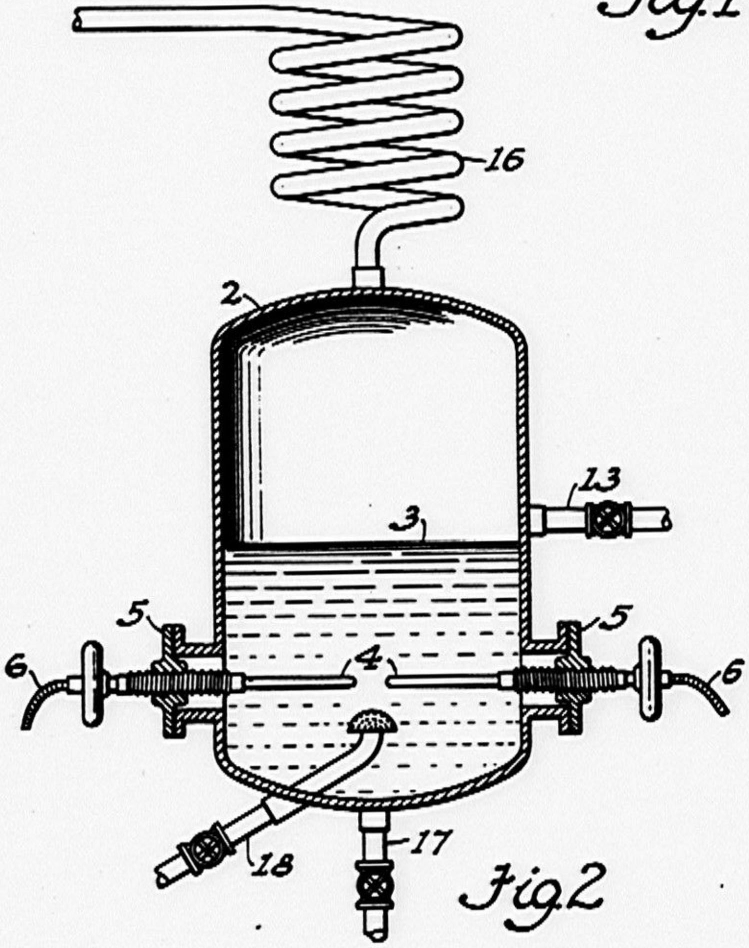

Fig. 2

Inventors
Frederick W. Sullivan, Jr.
Fred F. Diwoky
By Bruce K. Brown Atty.

Patented Dec. 5, 1933 **1,938,547**

UNITED STATES PATENT OFFICE

1,938,547

LEADIZED GASOLINE

Frederick W. Sullivan, Jr., Hammond, Ind., and Fred F. Diwoky, Chicago, Ill., assignors to Standard Oil Company, Chicago, Ill., a corporation of Indiana

Application March 21, 1932. Serial No. 600,222

12 Claims. (Cl. 204—31)

This invention relates to processes of preparing hydrocarbon compounds of lead and it comprises a process wherein a hydrocarbon, for example hexane, is dissociated by the action of an electric spark to liberate free alkyl radicals such as methyl, ethyl, propyl or higher alkyl radicals, and these reaction products are caused to combine with lead whereby lead alkyl compounds are formed.

Hydrocarbon compounds of lead, such as tetra ethyl lead, have been known for a number of years and are, at the present time, used in great quantities as anti-knock substances in motor fuels. In general, these lead alkyl compounds are prepared by reacting an alkyl chloride or bromide with a lead alloy such as an alloy of sodium and lead. There are a number of other ways of making lead alkyl compounds all of which in general require the use of an alkyl chloride and an expensive metal such as sodium or magnesium or alloy thereof.

We have now found that hydrocarbon compounds of lead can be prepared directly from metallic lead and hydrocarbons; we find that it is possible to dissociated hydrocarbons by means of an electric spark whereby free alkyl radicals are formed and that these free radicals will combine with lead.

Our process is characterized by providing a high tension electric arc of the order of 25,000 volts, between lead electrodes and maintained either in a body of liquid hydrocarbon or in a body of hydrocarbon vapors, the hydrocarbon being one which, when subjected to the action of the arc, dissociates to give free alkyl radicals. Thus, under the conditions of our invention, such dissociation of the hydrocarbons occurs in the region of the arc and in contact with metallic lead, either in fine particle form or as a lead "fog", as will be more fully explained. These conditions, we find, are therefore appropriate to the immediate formation of significant quantities of alkyl compounds of lead by reaction of the lead with the free alkyl radicals. The lead compounds thus obtained can be recovered either in a pure state or in solution in the hydrocarbon liquid in which they are formed.

On the appended single sheet of drawings we have indicated two advantageous ways of carrying out our process. In Figure 1, we have diagrammatically shown suitable apparatus for carrying out our process when the decomposition occurs in vaporized hydrocarbon material. In Figure 2 we have shown an advantageous form of apparatus for use in our process when it is desired to carry out the decomposition within a body of liquid hydrocarbon.

Referring more particularly to Fig. 1 a reaction vessel 1 contains a body of liquid hydrocarbons, such as hexane or heptane. Above the liquid level of the hydrocarbon, shown at 3, we provide lead electrodes 4. The lead electrodes pass through the sides of the vessel 1 through insulating bushings 5. The electrodes are connected to a source of high voltage alternating current by current conductors 6. A heating coil 14 is provided in receiver 1 to facilitate vaporization of the hydrocarbon therein. At the top of the reaction vessel there is provided an outlet conduit 7 for reaction products. Conduit 7 leads to condenser 8 terminating in a receiver 9. A vacuum pump, not shown, is connected to the receiver 9 by conduit 10 and condensed reaction products may be withdrawn from the receiver 9 through valved outlet 11. Cooling coils 12 are advantageously provided to maintain the contents of receiver 9 below a volatilizing temperature.

In the operation of the device shown in Figure 1, a hydrocarbon, which may be either liquid or gas, and which yields free alkyl radicals on electrothermal dissociation, is maintained in the reaction vessel 1. The system is preferably maintained under a reduced pressure, for example 1 to 5 millimeters of mercury absolute although higher pressures can be used. Under these conditions, a certain amount of liquid hydrocarbon will volatilize into the vapor space above the liquid level. Upon the institution of an arc between the lead electrodes, as by the application of a high tension current at about 25,000 volts thereto, volatilized hydrocarbon in the vapor space above the liquid level is dissociated. At the same time a fog or "mist" of lead particles is generated from the lead electrodes. The hydrocarbon dissociation products contain free alkyl radicals which immediately unite with the lead fog to form volatile lead-alkyl compounds. These volatile lead-alkyl compounds together with undissociated hydrocarbon vapors pass upwardly into conduit 7, condense in condenser 8, and collect in receiver 9. It is sometimes desirable although not essential, to provide receiver 9 with cooling means, such as refrigerator coils 12, in order to keep the lead compounds in the liquid state under the low pressure conditions employed. These lead compounds are very volatile substances.

Since the lead alkyl compounds are formed at the expense of the lead electrodes, the latter contributing to the lead fog described above, we advance the electrodes during the course of the reaction so that the arcing distance between them remains approximately the same. As a rule, the arc is about 1 to 20 millimeters long. That is to say, the ends of the electrodes are spaced apart about 1 to 20 millimeters. We maintain this distance throughout the reaction by advancing the electrodes to compensate for loss of lead from their arcing surfaces. This may be done in any convenient manner.

When using relatively high boiling hydrocarbon liquids such as hexane or heptane, it is sometimes advantageous to raise the temperature of the hydrocarbon liquid slightly to increase the vapor pressure of the liquid. This is easily done by passing a heating fluid through heating coil 14 within the reaction vessel. Likewise, if the heat of the arc causes too much volatization, a cooling fluid can be passed through coil 14 to restrain vaporization. The reaction product, which consists mainly of hydrocarbon compounds of lead, volatilized and condensed undissociated hydrocarbons, and uncondensible reaction products such as hydrogen, is cooled in condenser 8 to a temperature which insures the condensation of the lead-alkyl compounds as well as the undissociated hydrocarbon liquid. The uncondensible gases, such as hydrogen, pass out of the system through outlet 10.

The reaction products collected in receiver 9 consist very largely of lead-alkyl compounds dissolved in the condensed hydrocarbon fluid. When we wish to impart anti-knock qualities to a gasoline, we advantageously add to the gasoline the desired quantity of lead-alkyl compounds in solution in the hydrocarbon liquid as obtained in receiver 10. This obviates the necessity for recovering the lead compounds in a substantially pure state from the hydrocarbon liquid. The lead compounds can be recovered, however, in ways well known, for example by fractional distillation.

Our invention also provides a very convenient method of incorporating soluble lead compounds in gasoline itself. When we wish to do this we use gasoline as the hydrocarbon liquid in reaction vessel 1 and obtain a reaction product in receiver 9 consisting of condensed undecomposed gasoline in which the lead alkyl compounds are dissolved.

When using liquid hydrocarbons in the apparatus shown in Figure 1, the yield of lead-hydrocarbon compounds is a significant, though variable, quantity, depending, of course, upon such factors as the capacity of the apparatus, the rate of vaporization of the hydrocarbon liquid, the cross-sectional area of the lead electrodes and hence the amount of lead fog generated, the pressure in the system, and the particular hydrocarbon liquid chosen.

The product obtained in receiver 9 can be fractionally distilled to obtain the lead compounds in a substantially pure state and, when this is done, we advantageously recycle undissociated hydrocarbon. In this manner we are able to convert the larger portion of the original quantity of hydrocarbon material to lead compounds.

Cooler 12, in receiver 9 should be operated at such a temperature that no lead compounds collected in the receiver volatilize under the pressure conditions employed. When the pressure in the system is about 5 millimeters, the cooler should be at about —10° C.

When we wish to obtain a product consisting substantially of lead compounds free of undecomposed hydrocarbon material, we find it more advantageous to use normally gaseous hydrocarbons as the source of alkyl radicals. In this modification, we introduce gaseous hydrocarbons such as methane, ethane, propane or butane into the reaction vessel 1 through valved inlet 13. Since these substances are gaseous, the system need not be under reduced pressure and cooling coil 12 is unnecessary. Product obtained in receiver 9 consists substantially of lead-hydrocarbon compounds, and undissociated gaseous hydrocarbons leaving the receiver through outlet 10 can be recycled back to vessel 1. Hydrogen constitutes one of the reaction products obtained in the electrothermal dissociation and in the event that the hydrogen in the recycled hydrocarbon gases becomes too great, portions of the recycled gas can be vented to the atmosphere.

In Fig. 2, the reaction vessel 1 is provided with a valved inlet 13, lead electrodes 4, insulating bushings 5, and a reflux condenser 16. The reaction vessel is also provided with valved outlet 17 and a gas conducting pipe 18 which terminates at a point immediately beneath the adjacent ends of the electrodes. A body of hydrocarbon liquid 2 is maintained in the reaction vessel in such a manner that the liquid level 3 of the hydrocarbon is above the electrodes.

High tension current leads 6 are provided as in Fig. 1.

In the operation of the device shown in Fig. 2 a body of hydrocarbon liquid, for example, hexane or pentane, is maintained in reaction vessel 1 and an electric arc established between adjacent ends of electrodes 4. To assist in the initial establishment of the arc, we provide pipe 18 for the admission of a suitable gas such as hydrogen or ethylene. The hydrogen or ethylene, admitted to the space between the arcing surfaces of the electrodes, temporarily decreases the resistance to the passage of current across the arcing space which would otherwise exist due to the hydrocarbon liquid. The bubbles of hydrogen or ethylene thus enable the arc to start. After the arc has been established the flow of hydrogen or ethylene may be terminated and the arc will continue to function.

As soon as the arc is established, a certain amount of colloidal lead is formed in the body of hydrocarbon liquid as well as quantities of lead vapor. This is advantageous. The arc, when once established, dissociates the hydrocarbon liquid between the arcing surfaces with the liberation of free alkyl radicals. These combine at once with the lead in colloidal solution in the liquid or with the lead vapor from the electrodes. We also provide means to advance the electrodes so as to keep the arcing distance constant as the electrodes wear away.

The lead alkyl compounds formed remain dissolved in the body of hydrocarbon liquid and after a sufficient amount of lead is consumed we withdraw the reaction products through outlet 17. A reflux condenser 16 is provided to return hydrocarbon liquid which might be volatilized due to the heat of the arc.

As in the process described in connection with Fig. 1, we use a high tension current at about 25,000 volts or more. In the alternative method just described it is unnecessary to operate under a high vacuum. The process may be conducted under ordinary atmospheric pressure.

When carrying out our process in accordance with the showing in Fig. 2, all that is necessary is to maintain the arc within a body of hydrocarbon liquid. It need not be heated.

In our invention we can use any hydrocarbon liquid or gas, including the olefin hydrocarbons such as ethylene or propylene, so long as the hydrocarbon is one which can be dissociated by an electric spark to liberate free alkyl radicals.

Having thus described our invention, what we claim is:

1. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc within a body of hydrocarbon material of the type yielding free alkyl radicals on dissociation, immediately contacting the hydrocarbon dissociation products thus formed with metallic lead whereby the free alkyl radicals in said products combine with the lead to form hydrocarbon compounds of lead, and immediately removing the latter from the zone of the arc.

2. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc within a body of hydrocarbon gas of the type yielding free alkyl radicals on dissociation, immediate contacting the hydrocarbon dissociation products with metallic lead whereby the free alkyl radicals in said products combine with the lead to form hydrocarbon compounds of lead, and immediately removing the latter from the zone of the arc.

3. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc within a body of a liquid hydrocarbon of the type yielding free alkyl radicals on dissociation, immediately contacting the hydrocarbon dissociation products with metallic lead whereby the free alkyl radicals in said products combine with the lead to form hydrocarbon compounds of lead, and withdrawing the resulting solution of lead-hydrocarbon compounds from the zone of the arc after the desired quantity of such compounds has been formed.

4. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc between lead electrodes within a body of a hydrocarbon of the type which yields free alkyl radicals on dissociation thereby combining the free alkyl radicals thus formed with metallic lead and immediately removing the lead-hydrocarbon compounds from the zone of the arc.

5. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc between lead electrodes within a body of hydrocarbon gas of the type yielding free alkyl radicals on dissociation thereby combining the free alkyl radicals thus formed with metallic lead and immediately removing the lead-hydrocarbon compounds from the zone of the arc.

6. The process of preparing hydrocarbon compounds of lead which comprises establishing a high tension arc between lead electrodes immersed in a body of a liquid hydrocarbon of the type yielding free alkyl radicals on dissociation, thereby combining the free alkyl radicals thus formed with metallic lead and withdrawing the resulting solution of lead-hydrocarbon compounds from the zone of the arc after the desired quantity of such compounds has been formed.

7. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises dissociating a vaporized hydrocarbon by a high tension arc in the presence of metallic lead, the hydrocarbon being one which yields free alkyl radicals on such dissociation, whereby hydrocarbon compounds of lead are formed, and then condensing the reaction products.

8. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises dissociating a hydrocarbon gas by a high tension arc in the presence of metallic lead, the hydrocarbon being one which yields free alkyl radicals on such dissociation whereby hydrocarbon compounds of lead are formed, and then condensing the reaction products.

9. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises maintaining a body of a liquid hydrocarbon, and establishing a high tension arc therein in the presence of metallic lead to dissociate a portion of said hydrocarbon to form free alkyl radicals, whereby hydrocarbon compounds of lead are formed and dissolve in the main body of hydrocarbon liquid, the hydrocarbon being one which yields free alkyl radicals on such dissociation and withdrawing the resulting hydrocarbon solution of lead-hydrocarbon compounds from the zone of the arc after the desired quantity of such compounds has been formed.

10. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises dissociating a vaporized hydrocarbon by a high tension arc maintained between lead electrodes, the hydrocarbon being one which yields free alkyl radicals on such dissociation, whereby hydrocarbon compounds of lead are formed, and then condensing the reaction products.

11. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises dissociating a hydrocarbon gas by a high tension arc maintained between lead electrodes, the hydrocarbon being one which yields free alkyl radicals on such dissociation whereby organic compounds of lead are formed, and then condensing the reaction products.

12. The process of preparing a hydrocarbon liquid containing organic compounds of lead dissolved therein which comprises maintaining a body of a liquid hydrocarbon and establishing a high tension arc therein between lead electrodes to dissociate a portion of said hydrocarbon to form free alkyl radicals, whereby hydrocarbon compounds of lead are formed and dissolved in the main body of hydrocarbon liquid, the hydrocarbon being one which yields free alkyl radicals on such dissociation, and withdrawing the resulting hydrocarbon solution of lead-hydrocarbon compounds from the zone of the arc after the desired quantity of such compounds has been formed.

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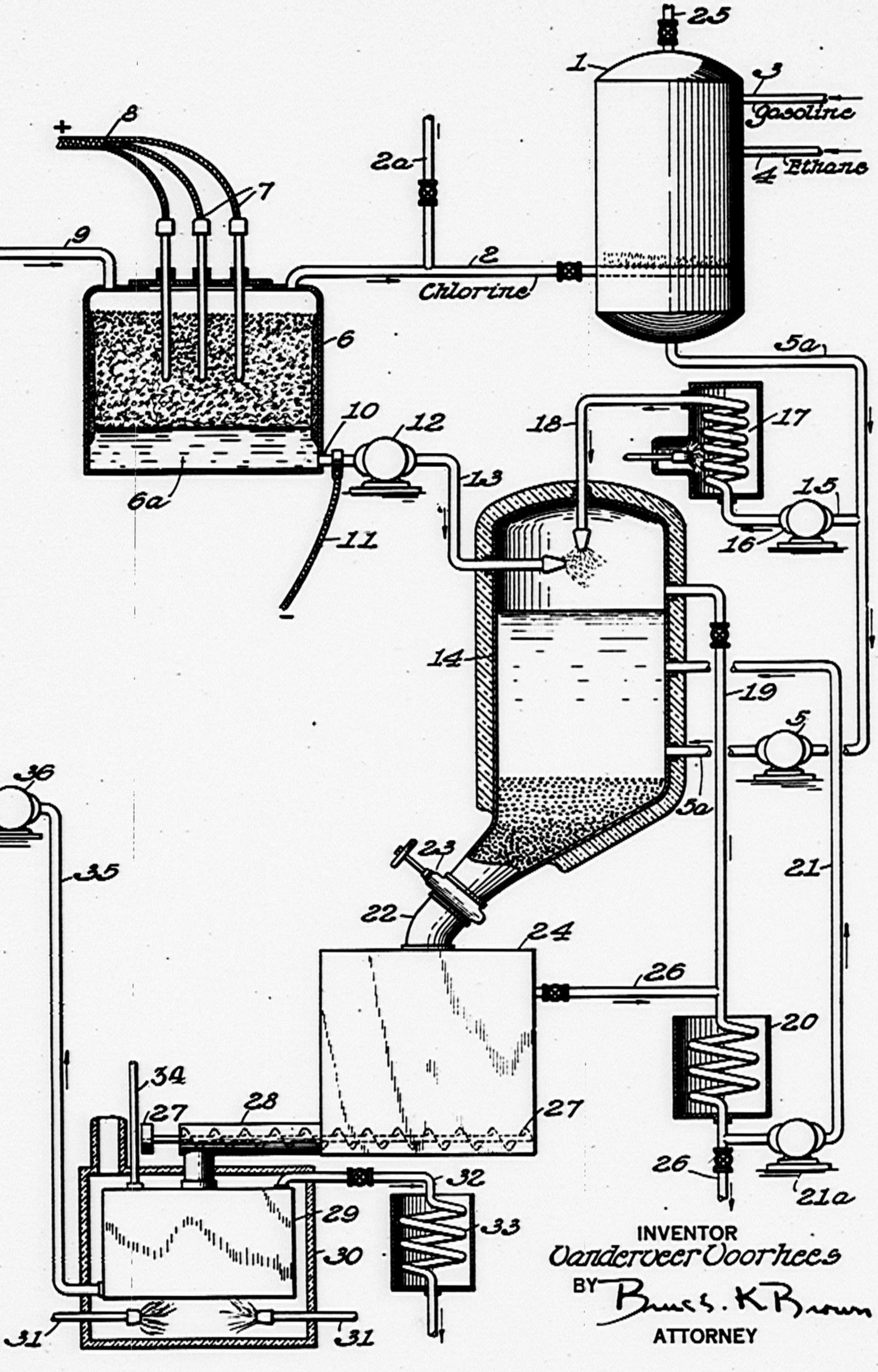
Sept. 18, 1934. V. VOORHEES 1,974,167

ANTIKNOCK GASOLINE

Filed Sept. 1, 1932



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UNITED STATES PATENT OFFICE

1,974,167

ANTIKNOCK GASOLINE

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Application September 1, 1932, Serial No. 631,386

5 Claims. (Cl. 44—9)

This invention relates to processes of preparing gasoline containing hydrocarbon compounds of lead and it comprises processes wherein gasoline is chlorinated with chlorine resulting from the electrolysis of fused sodium chloride in the presence of lead and the chlorinated gasoline reacted with lead-sodium alloy obtained in said electrolysis.

Tetra ethyl lead is a common ingredient in gasoline at the present time and is added to impart anti-detonating qualities to the gasoline. This "anti-knock" substance is customarily prepared by reacting ethyl chloride with a lead-sodium alloy. Its manufacture involves great industrial hazard and the product is expensive. The tetra-ethyl lead, made in the manner just described, is then incorporated in gasoline in certain definite proportions.

It is an object of the present invention to impart anti-detonating qualities to gasoline without the necessity of preparing tetra-ethyl lead or highly concentrated lead compounds. My invention has for its object the formation of hydrocarbon compounds of lead within the body of gasoline, in other words, in situ, so that I can dispense with the necessity for transporting tetra-ethyl lead and with the handling of this very poisonous material.

I have now found that gasoline can be chlorinated to such an extent that significant quantities of hydrocarbon chlorides are formed therein and that such hydrocarbon chlorides, while dissolved in the gasoline, will react with metallic lead, advantageously in the form of a lead-sodium alloy, to give me satisfactory quantities of hydrocarbon compounds of lead. Both operations are thus conducted in a large volume of gasoline and the hazards attendant the use of tetra-ethyl lead, or strong solutions thereof are avoided. Moreover, my process is less expensive and anti-detonating characteristics can be given the gasoline at less cost. It is not necessary that the pure substance, tetra-ethyl lead, be formed. So long as soluble hydrocarbon compounds of lead are formed within the body of the gasoline, as in my process, satisfactory anti-detonating properties are conferred on the gasoline. However, if desired, lead-hydrocarbon compounds having hydrocarbon radicals of the order of ethyl and propyl can be formed in the body of gasoline by dissolving therein, prior to chlorination, suitable quantities of ethane, propane, and butane, or unsaturated hydrocarbons having a like number of carbon atoms such as, for example, ethylene.

Advantageously in my process, and in the interest of economy, I make use of the products of electrolyzing fused sodium chloride in the presence of metallic lead. The electrolysis of fused salt, to make caustic soda is well known and is employed commercially. Ordinarily, in the operation of such electrolytic cells, chlorine is evolved at the anode and sodium plates out on a molten lead cathode. The sodium immediately alloys with the lead. When it is desired to make caustic soda the lead-sodium alloy is then reacted with water. In my process, however, I simply withdraw the sodium-lead alloy and react it with chlorinated hydrocarbons resulting from the chlorination of the gasoline by means of chlorine evolved from the same cell. Any unreacted lead-sodium alloy is ultimately returned to the electrolytic cell. Thus, my process produces a gasoline containing anti-knock substances derived from sodium chloride, metallic lead, electrical energy, and the gasoline itself. Since at no time do I have present quantities of pure or highly concentrated hydrocarbon compounds of lead, I have greatly reduced the industrial hazards in this art.

On the appended single sheet of drawings I have indicated in somewhat diagrammatic form a typical apparatus set-up which I find advantageous for use in my process.

Referring to the drawing, a chlorination vessel 1 is supplied with a chlorine inlet 2 which terminates in a plurality of distribution orifices within the vessel. Gasoline to be chlorinated enters the vessel by way of pipe 3 and provision for the introduction of ethane, propane, butane, or unsaturated hydrocarbons such as ethylene and propylene is made at pipe 4. The chlorinated products together with gasoline leave vessel 1 by way of pipe 5 and enter reaction vessel 14 as shown.

An electrolytic cell 6 is provided with anodes 7 to which the positive terminal 8 of the electric circuit is connected. These anodes may suitably be made of graphite. Pipe 9 serves for the introduction of fused sodium chloride and lead. A body of molten lead 6*a*, serving as a cathode, lies along the bottom of the cell and the negative terminal 11 of the source of current is conveniently attached to metallic conduit 10 which conduit also permits the withdrawal of lead-sodium alloy from the bottom of the cell.

During electrolysis of the fused sodium chloride, chlorine is evolved at the top of the cell and leaves by way of conduit 2 to enter chlorination vessel 1. Sodium plates out on the body of molten lead cathode, alloys therewith, and the sodium-lead alloy is pumped through pipe 10, pump 12 and pipe 13 to the top of reaction vessel 14. As stated, chlorination products dissolved in gasoline flowing from vessel 1 are conducted to reaction vessel 14 by pump 5 and pipe 5*a*. A bypass 15, pump 16, vaporizer 17, and pipe 18 serve to by-pass a part of the chlorination product to the top of vessel 14 in close proximity to the outlet of pipe 13. This is to provide means to atomize the lead-sodium alloy so that it is finely divided before mixing with the chlorination product in reaction vessel 14. The by-passed gasoline containing chlorinated hydrocarbons is pumped under pressure by pump 16 and vaporized to some extent in vaporizer 17 so that when it meets molten alloy flowing through pipe 13 the alloy is atomized into fine particles.

The atomized particles of alloy drop through the body of chlorinated products in the reaction vessel. Since the molten alloy is at a high temperature, considerable heat is given up to the body of gasoline and chlorinated products and, in general, the temperature of the mixture undergoing reaction in vessel 14 is 300° to 600° F. Within reaction vessel 14, the chlorinated compounds are converted to hydrocarbon compounds of lead which are soluble in the gasoline present and the solution is withdrawn through valved pipe 19 and cooled by cooler 20. If desired, part of the cooled product may be recycled through the line 21 and pump 21*a* back to the reaction vessel 14 to control the temperature of the reaction.

Unreacted lead-sodium alloy collects at the bottom of the vessel 14. This excess alloy is conveniently withdrawn from time to time through outlet 22 provided with valve 23 and conducted to a separator 24. The separator is provided with a screw conveyor 27 to remove spent alloy and other separating reaction products. This arrangement is for the purpose of recovering any liquid products which flow out with the particles of lead alloy and such liquid products are withdrawn by way of pipe 26 and united with liquid product flowing out of the vessel by way of line 19. The conveyor 27 is caused to rotate so as to withdraw the lead alloy from separator 24 and conduct it to a fusion vessel 29 by way of pipe 28. Fusion vessel 29 is disposed within a furnace 30 advantageously heated by oil burners 31. Sodium chloride to be fused is fed into the fusion vessel through inlet 34. I advantageously mix some potassium chloride with the sodium chloride in order to reduce the temperature necessary to fuse the sodium chloride. Sodium chloride melts at 772° C. but the addition of potassium chloride reduces this somewhat. As stated, excess lead alloy is also conducted to the fusion vessel 29. Should this be contaminated to any extent with gasoline containing lead compounds, most of which, however, flows from vessel 14 by way of pipe 19, I provide an outlet 32 and condenser 33 to recover such desired reaction products by driving them off from the lead alloy undergoing fusion in vessel 29.

The mixture of lead alloy and fused sodium chloride is transferred from vessel 29 through pipe 35 and pump 36 to the cell inlet 9 and in this manner any unconsumed lead alloy can be easily recovered for re-use. The vessels may conveniently be arranged for gravity flow from vessel 29 to cell 6.

With the apparatus shown the process can be conducted continuously by allowing small amounts of unconsumed lead alloy to flow from the bottom of reaction vessel 14 into separator 24 and thence to fusion vessel 29.

In the operation of the apparatus illustrated, the electrolytic cell 6 is charged with fused sodium chloride advantageously mixed with some potassium chloride for the purpose stated above, and a layer of molten lead provided on the bottom of the cell. The cell is then operated in the usual way for preparing caustic soda. Generally the direct current is very high, usually about 8300 to 8500 amperes with a current density of 2800 amperes per square foot at the anode. Normally the voltage is about 6.5 volts and the output of a cell about 3 feet x 6 feet having 4 anodes with an effective surface of about 3 square feet in all is roughly 495 pounds of chlorine per 24 hours. Advantageously I provide a number of such cells feeding the chlorine to a chlorine main which in turn supplies a plurality of chlorination vessels. The electrolytic cell as stated, is initially provided with a molten lead cathode which, as the electrolysis proceeds, is converted to lead-sodium alloy. Portions of the alloy are withdrawn from time to time to provide reagent in reaction vessel 14 and, as described, excess alloy is returned to the cell. As lead is consumed in the reaction forming lead compounds, further quantities of metallic lead can be added to the fusion vessel to make up for that consumed.

Virgin gasoline can be used in my process and introduced into the chlorination vessel 1 but I find it more advantageous to chlorinate a cracked gasoline. The amount of gasoline flowing intermittently or continuously to the chlorination vessel will of course depend upon the amount of chlorine available in any unit of time. Generally I obtain a chlorine absorption of approximately .05 percent of the weight of the gasoline. Some hydrochloric acid gas may be evolved and this can be vented to the atmosphere by means of vent 25 shown at the top of the chlorination vessel.

It is usually unnecessary to add hydrocarbons such as ethylene and this is especially true when cracked gasoline is used. However, I find it advantageous some times, depending upon the character of the hydrocarbons in the gasoline, to introduce quantities of normally gaseous aliphatic hydrocarbons by way of pipe 4. If the character of gasoline being treated is such that it is difficult to convert small portions of it rapidly to hydrocarbon chlorides, the introduction of ethylene or propylene may be resorted to and I can thus prepare lower aliphatic chlorides within the body of gasoline. Generally speaking, quantities of material and size of apparatus should be so proportioned that the final product obtained from pipe 26 contains about 0.2 to 0.4 percent of hydrocarbon compounds of lead. This will vary with the nature of the hydrocarbon compounds, some of them being more effective anti-detonating agents than others, but, chlorination should proceed to such an extent that subsequent conversion of chlorinated products by reaction with the lead alloy will be sufficient to give me a gasoline containing approximately 0.2 to 0.4 percent of dissolved hydrocarbon compounds of lead. However, higher concentrations of lead compounds may be obtained and the product may be used as blending stock. Thus a stock containing 4% of lead as lead compounds may be diluted with about 10 volumes of untreated gasoline to give a satisfactory antiknock fuel. The extent of chlorination governs the amount of lead compounds ultimately formed

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since the reaction with lead alloy is practically quantitative.

Lead alloy leaving the electrolytic cell is at a high temperature. Pure lead melts at about 620° F. If the lead contains 13 parts of sodium alloyed therewith the melting point of the alloy is about 624° F. I find it desirable to so regulate the electrolysis of the fused salt that the lead alloy contains from 5 to 15% of sodium. Such an alloy will leave the cell at a temperature at least high enough to melt the alloy (about 625° F.) and generally considerably higher since the fused salt is at a temperature of about 770° C. or somewhat less if potassium chloride is mixed therewith. When the molten alloy meets the volatized chlorination products by-passed through pipe 15, the alloy may be chilled somewhat and, at the same time, quickly atomized so that alloy falling into the body of chlorination products in vessel 14 is in a finely divided condition. This of course is desirable since it promotes reaction. Sometimes the lead alloy, after being atomized, remains molten. In this event, the atomized liquid mixes with the body of liquid hydrocarbon in the reaction vessel as a finely divided temporary suspension. However, as it settles down through the body of hydrocarbon, it solidifies in small particle form.

The heat derived from the hot lead alloy is usually sufficient to heat up the contents of the reaction vessel to reaction temperature, advantageously about 300° to 600° F. Further quantities of heat are also imparted to the contents of the vessel by the vaporizer 17. The reaction vessel should be large enough to give adequate time of contact between the particles of lead alloy and the liquid therein before the alloy collects at the bottom of the vessel.

The best temperatures to be maintained within the reaction vessel depend upon the composition of the alloy. When the content of sodium therein is low, in the neighborhood of 2 or 3%, somewhat higher temperatures, 600° F. or more, are necessary and, when the sodium content is high, about 15%, lower temperatures around 300° F., are satisfactory. However, if the sodium content of the alloy is too high the current efficiency of the cell will be low. I therefore so regulate the action of the cell that I obtain an alloy containing from 5 to 15 percent of sodium. This gives me satisfactory current efficiency in the cell and also sufficient sodium in the alloy to obtain satisfactory reaction temperatures and reaction rates within the reaction vessel 14. An alloy containing 5 to 15 percent of sodium will react satisfactorily at any temperature within the range of 300° F. to 600° F.

Should the sodium content of the alloy be below 5 percent, and thus require a reaction temperature of over 600° F. in the reaction vessel, I can supplement the heat derived from the molten alloy by by-passing and vaporizing more chlorination products through pipe 15. Sometimes it is desirable to operate at reaction temperatures such that the lead alloy retains its molten state throughout the reaction. This of course requires that the alloy shall not be chilled below solidifying temperature during the atomizing stage and I accordingly by-pass and vaporize more chlorination products when this is desired. In such case the molten alloy which has not been consumed collects in the bottom of the reaction vessel and is run off through outlet 22. Such molten alloy contains practically no liquid reaction products and I can therefore dispense with the separator and conveyor arrangement, and conduct the molten alloy directly to the fusion vessel for recovery and re-use.

Since the reaction vessel is maintained at a high temperature, in excess of the boiling point of the hydrocarbon fluids therein, it is necessary that the whole system be kept under pressure sufficient to maintain the hydrocarbon fluids in the liquid condition. This pressure varies with the temperature and may be about 300-800 pounds per square inch.

It will of course be obvious that other arrangements of apparatus can be employed together with numerous details to facilitate the handling of the materials. I therefore do not wish to be bound by the specific form of apparatus illustrated.

The gasoline product obtained in my process contains 0.2 and 0.5 percent or more of dissolved hydrocarbon compounds of lead the hydrocarbon radicals of which have been directly derived from the gasoline itself. This product is highly antidetonating and is superior to gasoline made by the addition of antiknock substances. Although I am not prepared to state the exact nature of the lead-hydrocarbon compounds in my gasoline I believe that they are more complex in chemical structure than the simple alkyl compounds and their hydrocarbon radicals contain in general at least 4 carbon atoms. They are probably mixtures of lead derivatives of various isomeric liquid hydrocarbons having from 4 to 7 carbon atoms.

Although as stated above, conversion of chlorinated hdrocarbons to lead compounds appears to be substantially quantitative, small amounts of chlorinated hydrocarbons may be left in the final product and assist in the antidetonating action of the lead when the fuel is burned in the engine. The quantity of lead compounds formed in the operation is determined by the extent of chlorination. I may discard part or at times, all of the chlorine through valved line 2*a*, to control the extent of chlorination. For this reason I have not specifically claimed any particular proportional composition of gasoline and lead hydrocarbon compounds.

I also find that my gasoline product is practically sulfur free because of the fact that such sulfur compounds as may be present in the gasoline are taken out by the lead alloy. Thus the alloy not only converts chlorinated hydrocarbons to lead compounds but also desulphurizes the gasoline. This is particularly advantageous since the raw gasoline entering the system need not be highly desulfurized. I find that in my process where the chlorinated hydrocarbons are dispersed in a large volume of gasoline, they may be converted into lead derivatives at higher temperatures without decomposition of the resulting derivatives.

Having thus described my invention, what is claimed is:

1. The process of preparing gasoline containing lead compounds which comprises electrolyzing fused sodium chloride in the presence of lead to form chlorine and a lead-sodium alloy, reacting the chlorine with gasoline to form chlorinated hydrocarbons therein, and treating the gasoline containing the chlorinated hydrocarbons with the lead-sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead.

2. The process of preparing gasoline containing lead compounds which comprises electrolyzing fused sodium chloride in the presence of lead to form chlorine and a lead-sodium alloy, reacting the chlorine with gasoline containing dissolved, normally gaseous, hydrocarbons to form chlorin-

ated hydrocarbons therein, and treating the gasoline containing the chlorinated hydrocarbon with the lead-sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead.

3. The process of preparing gasoline containing lead compounds which comprises electrolyzing fused sodium chloride in the presence of lead to form chlorine and a lead-sodium alloy, reacting the chlorine with gasoline to form chlorinated hydrocarbons therein, treating the gasoline containing the chlorinated hydrocarbons with the lead-sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead, separating the gasoline containing the hydrocarbon compounds of lead from unreacted lead-sodium alloy, and uniting the latter with further quantities of fused sodium chloride to be electrolyzed.

4. The process of preparing gasoline containing lead compounds which comprises electrolyzing fused sodium chloride in the presence of lead to form chlorine and a lead-sodium alloy, reacting the chlorine with gasoline containing dissolved, normally gaseous, aliphatic hydrocarbons to form chlorinated hydrocarbons therein, treating the gasoline containing the chlorinated hydrocarbons with the lead-sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead, separating the gasoline containing the hydrocarbon compounds of lead from unreacted lead-sodium alloy, and uniting the latter with further quantities of fused sodium chloride to be electrolyzed.

5. The process of preparing cracked gasoline containing lead compounds which comprises chlorinating the gasoline with chlorine, derived from the electrolysis of fused sodium chloride in the presence of lead, to form chlorinated hydrocarbons and treating the chlorinated hydrocarbons with the lead-sodium alloy resulting from such electrolysis to form hydrocarbon compounds of lead.

VANDERVEER VOORHEES.

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UNITED STATES PATENT OFFICE

2,012,356

METALLO ORGANO DERIVATIVES

Sol Shappirio, Washington, D. C.

No Drawing. Application November 18, 1932,
Serial No. 643,303

11 Claims. (Cl. 44—9)

This invention relates to metallo organic compounds and to processes of making the same, and particularly to products especially adapted for internal combustion engines containing such metallo organic substances. This application is a continuation in part of application, Serial No. 111,886, filed May 26, 1926, entitled "Process of making metallo organic compounds and compositions containing the same".

Recently there has been a marked development in special fuels best known perhaps in connection with the introduction of such compounds as lead tetra ethyl into low compression fuels. Other analogous or related metallo compounds have also been suggested. However, these metallo organic compounds have usually been made by more or less classic and long known processes with but slight modification. The preparation of these compounds by such processes on commercial scale for industrial use have given rise to great difficulties in connection with the handling of the ingredients utilized and more particularly the products sought. Such compounds as lead tetra ethyl have a highly poisonous character and even in very small amounts cause untold suffering and ultimately death in the case of workers who have come into contact with it during manufacture. Such happenings have been largely responsible for the agitation recently against both the sale and manufacture of these compounds and fuels containing them.

Among the objects and advantages of the present invention is the preparation of such metallo organic compounds by processes which eliminate to a large extent many of the hazardous elements of such manufacture, and enable the materials to be handled without danger, and in fact avoid many of the steps that have heretofore been found to be necessary in their preparation.

Basically this invention involves the treatment of oils of hydrocarbon character or their derivatives so that they are converted into compounds, in part, or to a limited extent, which upon further treatment are readily converted into metallo organic derivatives. It is thus seen that the metallo organic derivatives are prepared directly in the fuel or analogous material, and preferably from constituents of the fuel or analogous material itself, without the addition of extraneous hydrocarbon sources. But this invention is not limited to such operation, since there may be added to the gasoline or other material treated, extraneous substances either for the purpose of forming the metallo organic substances within the fuel, or as a partial source for such metallo organic substances, some of the latter also being derived from the fuel or analogous material.

As the source of hydrocarbon material that may be used there may be mentioned gasolines produced by various processes and other hydrocarbon products that have fuel value. Such gasolines may be those from straight distillation processes, from cracking processes either in the presence or absence of air, from natural gas, etc. It is desirable to have present some of the lower members of the paraffin series, and consequently gasoline containing such lower paraffins such as ethane, propane, and butane are particularly desirable for treatment in this process. Further gasolines containing aromatics are also highly desirable since these are readily converted into the desired metallo organic compounds referred to in more detail below. Further, gasolines and related materials that have been produced during cracking or analogous heat and pressure treatment in the presence of air are highly advantageous, and more particularly when in such processes the gasolines produced contain even a small amount of oxygen containing substances, such as the ethers, since such oxygen containing substances often act catalytically in the processes of converting some of the described intermediate substances into metallo organic compounds.

When certain desired low members of the paraffin series are not present in the gasoline or other material to be treated, they may be added to the same; and of course similar considerations apply to any other constituents that may be desired in the final products. However, instead of converting the constituents of the gasoline or analogous material directly into the intermediate that is to be converted into the metallo compounds, there may be added to the gasoline or similar material, a substance which will be converted into the metallo compound desired, the gasoline remaining substantially as it was before the conversion and the metallo compunds being derived solely from the added substances. Any combination of these steps may be utilized depending on the products desired.

Among the metals and non-metals that may be introduced into gasoline by the processes herein set forth there may be mentioned lead, tin, silicon, antimony, arsenic, phosphorus, mercury, thallium, etc. In describing the processes illustrations will be given showing the application to the introduction of lead into the gasolines, but this is done merely as illustrative of what may be accomplished by the invention, since derivatives of the stated and other metals and non-metals may be readily prepared by strictly analogous processes. Lead compounds are, however, those generally in use and the illustrations are accordingly drawn with this in mind. But it will be obvious that various changes may be made in this disclosure by those skilled in the art without departing from the scope and spirit of this invention.

Primarily this invention is concerned with the preparation of the desired metallo organic compound in the fuel or analogous material, so that the metallo organic compound does not have to be separately handled. The fuel may be present in a more limited quantity so that a concentrated metallo organic containing product is obtained which may be obtained by distilling off the hydrocarbon products, or which may subsequently be diluted with more fuel or gasoline or related substance to the concentration desired. Or the metallo organic substance or substances may be prepared in the gasoline or analogous material in a very dilute condition so that further dilution is unnecessary, and the special fuel is directly produced without more.

In many of the reactions of making metallo organic derivatives, the presence of catalysts have been found to be highly desirable. Such catalysts usable herein are particularly referred to as Grignard catalysts since the catalysts generally used for catalyzing the Grignard reactions may be used herein. As such catalysts there may be mentioned primary, secondary, tertiary amines and their alkyl addition products, such amines being aliphatic, aromatic or heterocyclic derivatives. Examples are aniline, dimethylaniline, carbazol, phenyl hydrazine, quinoline, etc. Other catalysts that may be used are ether, ammonia, esters, ethers in general, etc., etc. Some of these compounds may exist in the gasoline or other material undergoing treatment, so that the addition of such catalysts is not specifically necessary. On the other hand the catalysts may be added to the gasoline or analogous material, and when they are so added they may be selected from the named or other substances with regard to their value in fuel compositions. Some of the simpler ethers, and amines particularly of the aromatic series may be thus chosen.

As pointed out above, the process herein disclosed involves two essential steps, namely, first the treatment of the gasoline or analogous material to render it capable of being converted in part into metallo organic compounds, or the addition to the gasoline or analogous substance, of compounds that may be readily converted into such metallic derivatives, and second the formation of the metallo compounds therefrom. The metallic derivatives which it is desired to produce are the alkyls and aryls, such as lead tetra ethyl and antimony phenyl, and related substances. But it will be understood that when the term metallic alkyl or aryl is used, it is intended to cover the non-metallic compounds such as those of phosphorus and other non-metals, which form analogous compounds of similar properties by parallel processes.

As exemplifying one method of applying this invention, the following is given. Gasoline is given a slight treatment with halogenation agents in order to halogenate a portion thereof to introduce, say 10% of halogen. The halogens used are preferably chlorine or bromine, and the usual processes of carrying out such halogenation are used. The presence of water is desirably avoided by using dry materials. The process is preferably carried out so that mainly monohalogen derivatives are formed. It is unnecessary to halogenate but a small portion of the gasoline since the remainder will act as a solvent for the halogen derivatives formed both in this step and in subsequent operations. However, as explained above, the gasoline or analogous material may be rather completely halogenated to the mono derivatives and either then diluted, or first converted into the metallo derivatives and subsequently diluted. In the latter event a concentrate of the metallo derivatives will be obtained, or the metallo derivatives obtained without any diluting substances present.

As stated the halogenation should be carried out to give a maximum of mono halogen derivatives and a minimum of higher derivatives. Where the latter are present in undesirable amount, they may be first separated before proceeding to the formation of the metallo derivatives, as by distillation. Among the halogenation processes available and well known in the art for producing the halogenation of the gasoline or related material in accordance with the present disclosure, reference may be made to the U. S. patents to Brooks, 1,191,916; Blanc, 1,248,065; and Sparre, 1,379,367, which patents disclose methods that may be employed for halogenating gasoline. The procedures of these patents or other patents well known in the art, and well known methods of the art for producing halogenated products and particularly from gasoline and related material may be employed.

Instead of proceeding in this way, the desired amount of a halogen derivative may be introduced into the gasoline. For example, ethyl chloride, methyl chloride, propyl bromide, etc. may be added to the gasoline or analogous material in the desired proportion to yield the requisite amount of metallo derivatives in subsequent operations.

It should be noted that in the final fuels containing such metallo derivatives, they need be present in but small amount only, for example less than one percent, and even less than one-quarter of one percent. Consequently whether the halogen derivatives are formed in the gasoline or analogous material directly by halogenation processes, or whether desired halogen compounds are added thereto, they need be present in but small quantities if desired, for example in such quantities as will yield the requisite proportions of metallo compounds by further reaction desired in the final fuel. But it is found to be preferable to produce the halogen content above that finally desired, convert such halogen content into the metallo derivatives, either partially or completely, and having thus produced a concentrated metallo content in the fuel composition, dilute this when desired to the final composition sought.

Further, as indicated above, it is well to choose gasolines or analogous material that contain a substantial proportion of the lower paraffins, or to enrich the gasolines with such lower paraffins or their derivatives, in order to form a preponderant amount of the lower metallo alkyls. This is well taken care of in view of the fact that in most of the reactions used for forming the metallo derivatives, the lower paraffins react much more readily than do the higher ones, so that it is possible to control the reaction and produce more of the lower metal alkyls, than of the higher ones. Instead of enriching a gasoline with lower paraffins, it may be enriched with aromatics such as benzene, toluene, xylene, etc. when these are not present, and if they are desired, in order to form the metal aryls in the fuel.

Having thus produced a halogen derivative content in the gasoline or analogous material of desired amount, the solution preferably without water present, is subjected to any well known process for converting the halogen derivatives into metal compounds, such conversion taking place directly in the fuel itself, any excess of the latter not halogenated forming a solvent. When desired, some additional fuel may be added as solvent, or extraneous solvents may be added both for their solvent purposes and also for action as catalysts.

Two methods particularly may be used at this point for a conversion of the halogen derivatives into metal compounds. The halogen compounds may be converted into their corresponding magnesium compounds (Grignard reagents) and then reacted with heavy metals, their alloys, or compounds such as salts, in order to form the metal derivatives; or the halogen derivatives may be treated with metallic substances, their alloys, or compounds to form the desired metal derivatives in the fuel. These reactions are preferably carried out in the presence of catalysts and sometimes in the presence of reducing agents in order to eliminate certain side reactions or byproducts, and desirably the presence of water in substantial amount is avoided. Several examples will be given, it being understood that these are merely for purposes of illustration, and are not limiting in any way, since the invention is capable of application in a wide variety of ways.

The halogen derivatives contained in the fuel substance is subjected to the action of magnesium desirably in the form of wire or band. Such magnesium may be either immersed in the fuel mixture, or the latter may be circulated through a bed of magnesium until the halogen content of the fuel substance has been converted into the magnesium compound in accordance with the following equation:

$$RCl+Mg=RMgCl$$
$$RBr+Mg=RMgBr$$

where R stands for either an alkyl or an aryl radicle. In carrying out the conversion of the halogenated material into the metal Grignard reagent, it is not necessary to convert all of the halogen compounds into magnesium compounds, but any desired proportion of the halogen derivatives may be allowed to remain therein within the limits sought, since in the special fuels containing such compounds as lead tetraethyl it is desirable to have present halides, such as the chlorides and bromides, in the final fuel as it is consumed in the internal combustion engine for example in order to prevent the deposition of lead in the engine. Consequently the process can be so carried out that considerable quantities as desired of the halides remain in the fuel. Or when making metallo organic concentrates as described above, a considerable portion or all of the fuel remaining unconverted into the metal derivatives, may be in the form of halides which can then be diluted with more fuel to obtain the final product ready for use.

Among the methods which may be employed for producing the desired metallo organic derivatives, specific reference may be made to the process abstracted and referred to in Chemical Abstract, vol. V, at page 2639, and particularly the method therein disclosed for producing these metallo organic combinations in the presence of such organic material as ligroin, benzene, etc.

The Grignard type of reagents having been prepared as set forth above, it is then treated to form the desired metallo compound by reaction with desired heavy metal or non-metal compounds. For example, in the preparation of lead tetraethyl the alkyl or aryl magnesium halides contained in the fuel mixture, may be reacted with a lead salt such as the chloride to form the corresponding lead compounds. One equation illustrating the reactions that take place in such processes is the following:

$$PbCl_2+4C_2H_5MgBr=Pb(C_2H_5)_4+MgBr_2+2MgBrCl+Mg$$

Or again the following will illustrate the reaction:

$$2PbCl_2+4C_2H_5MgBr=Pb+Pb(C_2H_5)_4+2MgCl+2MgBr_2$$

These equations are merely given to illustrate several reactions that may take place and being theoretical only are not intended to be limiting. Other metals yield corresponding compounds as well of the alkyls as of the aryls.

In view of the fact that the lower paraffins more readily form the metal alkyls, the reaction can be controlled so that more of the lower metal alkyls are present than are the higher alkyls; and a similar control can be exercised in the halogenation reactions to limit the halogen derivatives formed largely if not entirely to the lower paraffins; or similarly this result may be reached by the addition of halides to the desired gasoline or fuel, using the lower alkyl halides only.

Other metal salts or compounds may be used to obtain similar reactions and the production of corresponding metal organic derivatives in the fuels. And analogous reactions may be produced with non-metal compounds. As pointed out above, catalysts are desirably used in most of these reactions, and as such the catalysts may be added compounds, particularly those that have a value in the final fuel composition, or merely solvents such as ether which also exert a catalytic influence. When as frequently happens, some of the byproducts are insoluble in the fuel, they may merely be filtered out. But if desired, they may be washed out with water or other washing solutions.

Instead of making the magnesium derivatives from the halides, the later may be used directly without forming the magnesium derivatives in the presence of magnesium, ether or other appropriate solvents and catalysts. The intermediate step of forming the magnesium is thus eliminated. Further in place of using magnesium, zinc may be used to form intermediate compounds which may be used in an analogous manner.

And still further, the methods of reacting the halides with lead, lead alloys, or lead compounds may also be used without forming magnesium or corresponding derivatives at all. For example, the fuels containing the halides may be treated with lead or lead alloy (sodium alloy containing from about 6 to 20 percent of sodium) in the presence of reducing agents and catalysts. In these reactions the temperatures are generally maintained between 20 and 36° C. unless pressure is employed in which case the higher temperatures may be used. When not using pressure, the upper limit of temperature is largely controlled by the boiling points of the fuel ingredients. And the reaction products are often washed with strong alkali to improve the yields of the desired metal alkyls. When lead or other metals, or their alloys are used, these are preferably used in a more or less finely divided condition to increase the surface contact. The reduction reaction may be effected due to the reaction of the sodium in the alloy for example, with a small amount of water added to the materials undergoing treatment. But reduction may be carried out in any other desirable way.

Depending to a large extent on the amount of halides present undergoing reaction in such compositions, the temperature control will vary. If large quantities of the halides are present, the temperature may rise so that cooling is necessary to keep it within the prescribed limits. But where the amount of halides is small in comparison with the solvent or fuel elements present, heating may be required to a limited extent to raise the temperature.

In any reactions where the halides or the magnesium or similar Grignard reagents are used, and contacted with metals, their alloys or salts or other compounds, instead of adding the metals etc. to the fuel containing the halides, the fuel, etc. may be circulated through a chamber containing the metals, alloys or etc. until the desired degree of reaction has been obtained.

While a number of metal derivatives may thus be introduced into the fuel by being made directly therein, if desired after the production of one metallo organic compound, it may be converted either partially or wholly into other metallo compounds. And furthermore, if desired, instead of converting the Grignard intermediates produced as set forth above, or added to the fuel—for example, the Grignard reagents might be prepared in the usual manner and then added to the gasoline or other fuel—the latter may be converted into other derivatives than the metallo derivatives. The Grignards are well known as excellent intermediates for the synthesis of a number of organic compounds. So that they may be used as set forth herein to form a large number of organic compounds that have value in fuels. For example, by appropriate treatments they may be converted into alcohols, phenols, aldehydes, ketones, etc., and these reactions may be utilized so that a portion of the Grignard reagents are converted into the metallo derivatives and a portion into other organic compounds.

And further, many of the products may be made in the fuel from the halides by taking crude hydrocarbon materials that must be subjected to distillation or other treatment, or treatments, in order to obtain motor fuels from them, by adding the requisite materials to them to form the metallo compounds, or converting a portion of such crude materials into compounds that will form metallo derivatives, and then carrying out the distillation or other treatment, so that a gasoline or analogous material or fuel is obtained containing the metallo organic compounds that have been formed during the distillation or other treatment, which may or may not have been carried out under pressure.

Whenever desired, any material to be treated in accordance with the hereinabove set forth disclosure, may be dried if desired or necessary in order to remove any excess moisture that might be present and might interfere with the progress of the particular reactions sought in the production of specific compounds.

In carrying out the processes set forth herein, the reacting ingredients may be used in their theoretical molecularly equivalent proportions in accordance with the equations for the reactions as illustrated hereinabove, or an excess of any of the stated ingredients as compared with the material with which it reacts may be employed. Thus when the halogenated gasoline is reacted with the magnesium, the materials may be present in equivalent molecular amounts, but preferably the halogen derivative is used in excess. So too, when the Grignard reagent is formed, the material with which it is reacted is desirably used in slight excess.

Ordinarily as a result of this process, a complex mixture of organic derivatives of a stated metal, or metalloid is obtained, the derivatives, however, usually containing a number of closely related derivatives. Thus in a given instance, in making lead tetraethyl, there will also be present lead derivatives of other alkyls. And usually there is no necessity for isolating individual compounds, since these complex mixtures of such derivatives are desirably present in motor fuels for example, since they modify the action of each other having solvent action on each other, and hydrocarbons and their derivatives to yield homogeneity, smoother explosion curves, etc.

The metallo derivatives may be recovered as such by evaporation of hydrocarbons etc. from the reaction mixtures, or in any other desired way; or they may be diluted as with fuel ingredients for direct use.

While lead tetraethyl has been specifically referred to, antiknock metallo organo derivatives other than (and when desired to the exclusion of) lead tetraethyl, may be produced.

Having thus set forth my invention, I claim:

1. The method of making metallo organo derivatives which comprises treating gasoline to produce halogenated aliphatics therein convertible into metallo organo derivatives, and reacting such halogenated aliphatics under conditions to produce metallo organo derivatives with a metallic component to produce metallo organo derivatives the metallic component containing a metal of the anti-knock type, whereby the metallo organo derivatives possess anti-knock properties.

2. The method of making metallo organo derivatives which comprises treating a hydrocarbon oil with chlorine to form chlorides dissolved in the hydrocarbon oil, reacting such chlorides with magnesium to form Grignard bodies within the hydrocarbon oil, and then treating said Grignard bodies with a lead salt to form lead derivatives of the hydrocarbons.

3. A product containing a complex mixture of metallo organo derivatives the organic radicals of which correspond with the organic radicals of a group of hydrocarbons of a hydrocarbon oil.

4. A product containing a complex mixture of organic lead derivatives the organic radicals of which correspond with the organic radicals of a group of hydrocarbons of a gasoline.

5. A process of making anti-knock compounds which comprises halogenating petroleum hydrocarbons, and reacting such halogenated petroleum hydrocarbons with metallic reactants under conditions to form a mixture of anti-knock compounds of the metallo organic type, the metallic reactants containing a metal of the anti-knock type, whereby the metallo organo derivatives possess anti-knock properties.

6. A process of making anti-knock compounds which comprises metallizing a complex mixture of petroleum hydrocarbons containing halogen derivatives by reacting said mixture with metallic

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components to form metal containing anti-knock compounds from constituents of said mixture.

7. The method of making metallo organo derivatives which comprises treating a hydrocarbon oil with a halogen to form halides dissolved in the hydrocarbon oil, reacting such halides with magnesium to form Grignard bodies within the hydrocarbon oil, and then treating said Grignard bodies with a heavy metal compound to form heavy metal derivatives of the hydrocarbon the heavy metal compound containing a metal of the anti-knock type whereby the heavy metal derivatives of the hydrocarbon yield anti-knock properties.

8. The method of making metallo organo derivatives which comprises treating a hydrocarbon oil with chlorine to form chlorides dissolved in the hydrocarbon oil, and reacting such chlorides with lead alkali metal alloy to form lead derivatives of the hydrocarbons.

9. The process of preparing gasoline containing lead compounds which comprises reacting chlorine with gasoline to form chlorinated hydrocarbons therein, and treating the gasoline containing the chlorinated hydrocarbons with lead sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead.

10. The process of preparing gasoline containing lead compounds which comprises reacting chlorine with gasoline containing dissolved, normally gaseous, hydrocarbons to form chlorinated hydrocarbons therein, and treating the gasoline containing the chlorinated hydrocarbons with lead sodium alloy to convert the chlorinated hydrocarbons to hydrocarbon compounds of lead.

11. The process of preparing cracked gasoline containing lead compounds which comprises chlorinating the gasoline with chlorine, to form chlorinated hydrocarbons, and treating the chlorinated hydrocarbons with lead sodium alloy to form hydrocarbon compounds of lead.

SOL SHAPPIRIO.

III.

ADDITIONAL PAPERS DESIGNATED.

318 Filed Dec 22 1943

The United States of America, ss:

The President of the United States of America

(Seal)

To the Honorable the Justices of the District Court of the United States for the District of Columbia,

Greeting:

Whereas, lately in the District Court of the United States for the District of Columbia, before you or some of you, in a cause between Ethyl Gasoline Corporation plaintiff, and Conway P. Coe, and/or his Successor in Office, Commissioner of Patents, defendant, Civil Action No. 9986, wherein the judgment of the said District Court entered in said cause on the 27th day of October, 1942, is in the following words, viz:

This cause having come on to be heard and having been tried in open court and argued by counsel for the respective parties upon the pleadings and proofs adduced and submitted to the Court,

It is ADJUDGED this 27th day of October, 1942, that the complaint in this case be, and the same hereby is dismissed with costs against the plaintiff. MATTHEW F. Mc-
GUIRE, *Justice,* as by the inspection of the
319 transcript of the record of the said District
Court, which was brought into the United States Court of Appeals for the District of Columbia by virtue of an appeal, agreeably to the act of Congress in such case made and provided, fully and at large appears;
(Seal)

320 And Whereas in the term of October—, in the year of our Lord one thousand nine hundred and forty-

three,—, the said cause came on to be heard before the said Court of Appeals on the said transcript of record, and was argued by counsel:

(Seal)

On consideration whereof, It is now here ordered and—adjudged—by this court that the judgment—of the said District Court appealed from in this cause be, and the same is hereby, reversed, and that this cause be, and it is hereby, remanded to the said District Court for further proceedings in conformity with the opinion of this Court.

December 6, 1943.

321 You, Therefore, Are Hereby Commanded that such—further proceedings be had in said cause in conformity with the—opinion and judgment of this Court as according to right and justice and the laws of the United States ought to be had, the said appeal notwithstanding.

Witness the Honorable Harlan F. Stone, Chief Justice of the United States, the twenty-second—day of December—, in the year of our Lord one thousand nine hundred and forty-three.

JOSEPH W. STEWART,
Clerk of the United States Court of Appeals for the District of Columbia.

* * * * * * * * * *

322 Filed Feb 14 1944

Memorandum For The Clerk

Apart from the fact, as to whether or not the defendant based his claim "almost, if not entirely upon the proposition that the applicant had disclaimed his claims by reason of his failure to make the suggested claim"—this court having heard the case de novo on its merits fully and at length and prescinding now entirely from the suggested claim and the action of the Patent Office with respect thereto but having reference only to the cited patents is of the opinion that there is no invention, and so finds. This is as I view it what the Court of Appeals meant by its direction "the case will be remanded for findings and determination on the merits". The cited patents—Sullivan, et al; Voorhees, Shappirio and Midgley are all antecedent to the plaintiff's application.

Prepare findings of fact and conclusions of law consonant with this opinion.

MATTHEW F. McGUIRE,
Associate Justice.

February 14, 1944

* * * * * * * * * *

323 Filed Mar 29 1944

Findings of Fact Made Pursuant to Mandate of the United States Court of Appeals for the District of Columbia.

1. This is an action under Section 4915 R. S. (U. S. C., title 35, sec. 63) in which plaintiff, the assignee of the patent application of Earl Bartholomew, Serial No. 99,497, seeks a judgment which would authorize defendant, the Commissioner of Patents, to issue a patent including claims 1 to 12, inclusive, 15 and 16 of said application.

2. The application was filed in the Patent Office on September 4, 1936, and relates to method and means for improving engine operation and involves the use in the fuel of mixtures of some or all of the following lead alkyls, lead tetra-methyl, lead tri-methyl ethyl, lead di-methyl di-ethyl, lead methyl tri-ethyl, and lead tetra-ethyl.

3. Claims 1, 2, 3 and 4 of the application recite a method of operating an internal combustion engine. Claim 1 reads as follows:

324 1. The method of operating an internal combustion engine which comprises forming a combustible mixture of a motor fuel and air, compressing the mixture to a compression pressure greater than the critical compression pressure of the motor fuel and burning the mixture

together with a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

4. Claims 5 to 12, inclusive, of the application, recite a product comprising a motor fuel containing the anti-knock lead alkyl compounds. Claims 5 and 12 read as follows:

5. A product comprising a motor fuel and an anti-knock material having as its primary anti-knock component a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.

12. A product comprising a mixture of a motor fuel, a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetramethyl and lead tetraethyl present in relatively small concentrations.

5. Claims 15 and 16 of the application recite an antiknock composition comprising the lead alkyl compounds. Claim 15 reads as follows:

15. A fuel anti-knock composition comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl and lead tetra-methyl in a concentration not exceeding fifteen percent.

6. Claim 1 of the application specifies:

“* * * a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl.” Likewise claims 2, 5, 6, 7 and 8. None of these claims mentions either lead tetra-ethyl or lead tetra-methyl. Claims 2, 7 and 8 additionally require that “lead di-methyl di-ethyl” is the predominating one of the three named. Claim 3 specifies: “* * * a primary antiknock agent comprising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; and lead tetra-ethyl, the concentration of the lead being greatest in
the lead tri-ethyl methyl and lead tetra-ethyl.” Like-
325 wise claims 11 and 16. The only claims that mention
lead tetra-methyl are claims 4, 9, 10, 12 and 15.
Claim 4 specifies: “* * * a primary antiknock agent com-

prising a mixture of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl,; and lead tetra-methyl in a concentration not exceeding fifteen percent.'' Likewise claims 9, 10 and 15. Claim 12 specifies: '' * * * a primary antiknock agent consisting of lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl; * * *.'', being in that respect like the other claims that specify those three as the primary ingredients, and claim 12 further states: '' * * * and lead tetra-methyl and lead tetra-ethyl present in relatively small concentrations.'' Claim 12 is the only one of the claims that mentions all of the five lead alkyls, lead tetra-methyl, lead tri-methyl-ethyl, lead di-methyl di-ethyl, lead tri-ethyl-methyl, and lead tetra-ethyl. Claim 12 specifies that the lead tetra-methyl shall be present in relatively small concentration.

7. The patent to Midgley, No. 1,573,846, dated February 23, 1926, discloses the use of the lead alkyl compound, lead tetraethyl, $Pb(C_2H_5)_4$, and other alkyl compounds including the isopropyl and methyl compounds, as anti-knock agents in gasoline.

8. The patent to Sullivan, No. 1,938,547, dated December 5, 1933, discloses the method of combining the alkyl radicals, methyl, ethyl and propyl with lead to form lead alkyls and the use of mixtures thereof as anti-knock agents in gasoline.

9. The patent to Vorhees, No. 1,974,167, dated September 18, 1934, discloses the use of mixtures of lead hydrocarbon compounds in gasoline as anti-knock agents.

10. The patent to Shappirio, No. 2,012,356, dated August 27, 1935, discloses a process of making lead alkyl
326 compounds. The product is a complex mixture in which the lower lead alkyls predominate. The product is for use in gasoline as an anti-knock agent.

11. Claims 1, 2, 3 and 4 recite a conventional method of operating an internal combustion engine, and recite also the use of mixtures of certain lead alkyls as an anti-knock agent. The presence of such an anti-knock agent in the fuel does not cause the engine to operate by a method

which is different from its method of operation when using previously known anti-knock fuels.

12. Claims 1, 2, 3 and 4 are unpatentable.

13. The selection of the particular lead alkyls and the proportions thereof to be used in the fuel as recited in Bartholomew's claims depend on climatic conditions, the particular type of engine being used, and other factors. The selection is made as a result of experimentation.

14. There is nothing critical in the proportions of the ingredients, more or less indefinitely recited in Bartholomew's claims at bar.

15. None of Bartholomew's claims at bar recites anything amounting to an invention over the disclosures made in the patents to Midgley, Sullivan, Voorhees and Shappirio.

16. None of Bartholomew's claims at bar is patentable.

Conclusions of Law.

327 1. Plaintiff is not entitled to receive a patent containing any of claims 1 to 12, inclusive, 15 and 16 of the Bartholomew application at bar.

2. The complaint should be dismissed as to all of said claims.

MATTHEW F. McGUIRE
Justice.

* * * * * * * * * *

328 Filed Mar 29 1944

Judgment.

This cause came on to be considered further at this term pursuant to the Mandate of the United States Court of Appeals for the District of Columbia and thereupon, upon consideration thereof, it is this 29th day of March, 1944, Adjudged that the complaint be and it is hereby dismissed with costs against the plaintiff.

MATTHEW F. McGUIRE
Justice.

* * * * * * * * * *

329 Filed Apr 3 1944

Notice of Appeal to the United States Court of Appeals for the District of Columbia

Notice is hereby given that Ethyl Gasoline Corporation, plaintiff above named, hereby appeals to the United States Court of Appeals for the District of Columbia from the final judgment of this court dismissing the complaint which judgment was entered in this action on March 29, 1944.

H. FRANK WIEGAND
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New York 7, New York

A. K. SHIPE
1031 National Press Building
Washington 4, D. C.
Attorneys for Appellant.

Memorandum

April 7 1944

Bond of $250.00 for plaintiff on appeal approved and filed.

* * * * * * * * * *

Brief for Appellee

United States Court of Appeals

DISTRICT OF COLUMBIA

APPEAL No. 8760

ETHYL GASOLINE CORPORATION, APPELLANT

v.

CONWAY P. COE, COMMISSIONER OF PATENTS, APPELLEE

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

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E. L. REYNOLDS,
Of Counsel.

UNITED STATES
COURT OF APPEALS FOR THE
DISTRICT OF COLUMBIA

FILED JUL 10 1944

Joseph W. Stewart
CLERK

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United States Court of Appeals

DISTRICT OF COLUMBIA

APPEAL No. 8760

ETHYL GASOLINE CORPORATION, APPELLANT

v.

CONWAY P. COE, COMMISSIONER OF PATENTS, APPELLEE

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

BRIEF FOR THE COMMISSIONER OF PATENTS

INTRODUCTION

This is an appeal from the judgment of the District Court of the United States for the District of Columbia (182),[1] dismissing appellant's complaint (2–6) brought under Section 4915 R. S. (U. S. C., title 35, sec. 63), to authorize the Commissioner of Patents to issue to the appellant a patent containing claims 1 to 12, inclusive, 15 and 16 of the application of Bartholomew, Serial No. 99,497, which application is assigned to the appellant.

This case has previously been before this Court for review of the action of the District Court in dismissing the complaint on the round that the claims are unpatentable to Bartholomew because of the disclaimer involved in his refusal to make a claim suggested to him for the purpose of interference. The District Court held that, under the circumstances, the subject matter was to be treated as part of the prior art. This holding was reversed by this Court in a decision reported in 139 F. (2d) 372, and the case was remanded for "findings and determination on the merits." This Court, however, clearly did not hold, as stated in appellant's brief page 4, "that both the Patent Office and the District Court had erred in finding that appellant's claims were unpatentable because of his refusal to present the suggested claim." The holding was simply that, in determining the effect of the failure to adopt the proposed claim, its subject matter should not be treated as prior art. The Court stated that "The extent of the estoppel will depend, in the particular case, upon how the suggested claim is drawn and for what reason it is refused by the applicant," but expressed no opinion as to whether, under the circumstances of this case, such an estoppel existed. The holding was simply that the District Court had applied an incorrect principle in reaching the conclusion that there was an estoppel, and not that, if correct principles were applied, no estoppel could be found.

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[1] The numbers in parentheses refer to pages in appellant's appendix.

APPELLANT'S APPLICATION

The appellant's application (86) discloses an antiknock compound for internal combustion engines, comprising lead tri-methyl-ethyl, lead di-methyl di-ethyl, and lead tri-ethyl-methyl, to which may also be added lead tetramethyl and lead tetraethyl. The application indicates that considerable variations in proportions may be used to meet different operating conditions.

THE REFERENCES

The prior patents relied on are found on pages 155 to 176, inclusive, of appellant's appendix. They disclose the use of tetraethyl lead and of other lead methyl and ethyl compounds of varying volatility and mixtures thereof as antiknock agents for use in internal combustion engines.

APPEALED CLAIMS

The appealed claims appear on pages 3, 4, and 5 of appellant's appendix.

SUMMARY OF ARGUMENT

1. The Patent Office has not held the appealed claims patentable over the prior art.
2. The claims are extremely broad.
3. The use of the materials claimed is obvious in view of the prior art.
4. Claims 1 to 4 do not define a new method.
5. The evidence supports the holdings of the Trial Court.
6. The allowance of claims in other patents is not relevant here.

ARGUMENT

Before proceeding to a consideration of the merits of this case it is desired to point out that, although the appellant's brief states in no less than seven places that the Patent Office or one of its tribunals has held the appealed claims patentable over the prior art, there is no showing of record that any such holding has actually been made. The primary examiner (111) rejected all the claims of Bartholomew's application as unpatentable over the references now before the Court and later repeated this rejection (114). In a subsequent action (117) the claims were rejected as failing "to clearly define the invention disclosed," and at the same time a claim, differing from any claim here on appeal, was suggested for the purpose of interference. Bartholomew failed to present this claim within the time allowed and, in his next action (117), the examiner repeated his rejection on the ground that the claims were too indefinite to define the invention properly, and also rejected them on the disclaimer resulting from Bartholomew's failure to present the suggested claim.

It is evident from the examiner's statement, and was held by this Court in its former decision, above referred to, in this case, that the examiner, in making this rejection, treated the subject matter of the suggested claim as part of the prior art. So treated, this subject matter was, of course, a somewhat clearer anticipation than the prior patents and the examiner, therefore, evidently did not consider it necessary to urge the rejection on such patents further. He certainly, so far as the present record shows made no affirmative holding that the claims were patentable over the prior art.

The Board of Appeals (128) merely affirmed the examiner's decision. In its decision on a request for reconsideration (132) the Board said: "It was not intended to raise any new ground of rejection in our decision." On the basis of this statement the appellant, in his brief, page 3, says that the Board "held that it did not find any ground for rejection other than that stated by the examiner." The Board clearly did not make such a holding as this, but merely stated that it did not intend to add a new ground of rejection. It expressed no opinion as to whether or not such a ground would be valid.

It is thus clear from the record that neither the examiner nor the Board has expressed the opinion that the appealed claims are patentable over the references. The allowance of the suggested claim was not a holding that the present claims are patentable. This is obvious from the fact that the examiner (117) rejected these claims in the very action in which he made the suggestion. It is clear from the examiner's statement (125) that the suggested claim, in the examiner's opinion, properly defined the invention which Bartholomew was unsuccessfully seeking to define in the rejected claims. The examiner's statement (128) that the appealed claims could not be allowed because appellant has refused a contest "upon not patentably distinguishable subject matter" evidently means merely that, if the subject matter of the proposed claim be treated as prior art, the appealed claims are unpatentable thereover, and is not a holding that the appealed and suggested claims are not patentably different in scope, or that the allowance of one amounts to a holding that the others are patentable.

The statement in the appellant's brief (page 12) that "The patentability of the subject matter of the invention over the prior patents was conceded by the examiner when asserting that he was acting under Patent Office Rule 96, he formulated and presented the suggested claim" is rather surprising in view of the appellant's persistent contention that the suggested claim does not define any invention made by him. This contention is succinctly stated in the following portion of Bartholomew's testimony (65):

> Did the claim formulated by the examiner express your invention? A. No; it did not.

In view of this statement, it is not clear how the appellant can now contend that the allowance of the suggested claim has any bearing on the patentability of any invention made by him.

The appellant's argument seems to amount to an assertion that, if claims are rejected on one ground, this is an affirmative holding that there is no other conceivable ground of rejection which may be raised against them. This, of course, is not the case. It would be as logical to argue that a conviction for one crime is an affirmative holding that the criminal has never committed any other. The examiner merely urges what he considers the best grounds of rejection, and his failure to urge others does not mean that he has considered them and found them improper.

The contention that the Patent Office has held the appealed claims patentable over the art has been treated at some length because of the great emphasis placed on it in the appellant's brief, rather than because of its actual importance. The Trial Court, on the basis of the entire record before it, including the Patent Office actions, found that the claims were unpatentable, and the burden is on the appellant to show that this holding is wrong. He cannot, by merely filing an appeal, shift his burden to the appellee. The exact degree of the burden is not thought to be important since, as will appear, the claims are clearly unpatentable.

On page 13 of appellant's brief, the Findings of Fact in this case are criticized on the ground that they were "mechanically adopted by the Trial Justice over the objection of appellants." It is not clear what basis there is for the assertion that the adoption was "mechanical," but, in any case, the findings stand or fall on their own merits and it is not material how they were prepared (*Schilling* v. *Schwitzer-Cummins Co.*, Court of Appeals, D. C., decided March 31, 1944, 61 U.S.P.Q. 73).

NATURE OF INVENTION CLAIMED

The appealed claims are extremely broad and recite little more than the idea of using certain substances in an antiknock compound. No actual percentages of ingredients are given in any claim. Some of the claims make no statement whatever as to amounts, while others contain such indefinite statements as that an ingredient is present "in relatively small concentrations" or that the concentration of lead is "greatest" in certain of the ingredients. Still other claims specify "a concentration not exceeding fifteen percent," but, since they set no lower limit, this is indefinite. It is not suggested that any of the proportions thus broadly suggested are critical. Thus the application (120) merely states that a concentration less than 15 percent is "preferred"; as contrasted with the statement in appellant's brief, page 10, that "Bartholomew warns not to use over fifteen percent." Similarly, the application (102) states that the mixture "may have a relatively large concentration" of lead tetra-ethyl or lead tri-ethyl-methyl. It will also be noted that appellant, in his brief, does not seek to draw any distinction between the claims containing these indefinite allusions to proportions and those which contain none at all. Under the circumstances it is thought that if the claims are to be allowed, it must be solely on the basis of the character of the ingredients used and not upon the proportions since the appellant has not shown any specific proportioning to be critical nor even alleged that it is so.

The elaborate discussions contained in Bartholomew's application and testimony as to the manner in which knocking occurs in various cylinders and under various conditions are of little or no pertinence here. The claims are directed to the use of particular antiknock ingredients and not to theories of operation. In fact, although the theory advanced is concerned almost entirely with the relationship between the charges in the various cylinders, there is not one of the appealed claims which even suggests that the engine in which the fuel is used has more than one cylinder. Moreover, if the use of the compounds claimed was obvious in view of the prior art, it cannot be rendered inventive by theories as to why it is satisfactory (*Smith* v. *Hall,* 301 U. S. 216).

USE OF CLAIMED MATERIALS OBVIOUS

The use of lead compounds for preventing knocking is common, and the Midgley patent (157) discloses the use of tetra-ethyl lead as well as ethyl and methyl lead compounds for this purpose. Further, the Bartholomew application (87) states that "other lead compounds, particularly lead alkyl compounds, were generally known and some tried." The alkyl compounds include the various ethyl and methyl groups. It was also old prior to Bartholomew's alleged invention, to employ several different antiknock ingredients in the same fuel. Thus the Sullivan et al. patent (164, lines 80, 81; and lines 136 and 137) clearly contemplates the use of several different lead alkyl compounds at the same time, and among those mentioned are methyl and ethyl compounds. Similarly, the Voorhees patent (168) discloses the use of ethyl, propyl or butyl lead compounds, it being clearly suggested that several may be used at once. It is obvious that both Sullivan et al. and Voorhees contemplate the selection of such of these compounds as are considered desirable.

Further, the Shappirio patent (172) not only discloses the use of mixtures of lead derivatives of various alkyls, but recognizes the advantages flowing from such mixtures, as indicated by the following statement on page 4, column 2, line 21 et seq.:

> these complex mixtures of such derivatives are desirably present in motor fuels for example,

> since they modify the action of each other, having a solvent action on each other, and hydrocarbons and their derivatives *to yield homogeneity, smoother explosion curves, etc.* [Italics added.]

Obviously, therefore, there was nothing new in the conception that mixtures of lead alkyl compounds might produce a better result than the individual compounds.

Moreover, it was known prior to Bartholomew's work that each of the ingredients of his claimed lead compounds, namely, trimethyl ethyl, dimethyl diethyl, methyl triethyl, tetramethyl, and tetraethyl, could be used for antiknock purposes, and the degree of their effectiveness, at least under 100 percent vaporization conditions, was also known. This is apparent from the chart, Figure 4, of Bartholomew's application (110) and his testimony (46) relative thereto. The curve D of this chart is said to show the antiknock value of the substances at 100° vaporization, and this curve contains each of the claimed substances. With respect to this curve Bartholomew testified (46):

> This information with respect to the lead alkyls was generally known, perhaps not as accurately determined as here, but this particular quality of the alkyls was fairly well known.

The situation presented, therefore, is this: It was old to use mixtures of lead alkyls, including methyl and ethyl compounds, as antiknock agents, and it was also known that each of the particular compounds recited in the appealed claims possessed valuable antiknock properties. Bartholomew selected these particular compounds and now seeks a patent which would preclude others from using them together, in any proportion, whatever. Such a patent clearly could be sustained only if two conditions were present; first, that it would not occur to one skilled in the art to try these compounds together and second, that, in any proportions whatever, or at least in the proportions vaguely indicated in the more specific claims, they produce an unexpectedly superior result. It seems clear that neither of these conditions is satisfied.

It will be noted that, so far as appears, the separate compounds do not affect each other, but each produces its own effect, and the total result is the sum of these separate effects. Thus, one type of compound is found to be good under one set of conditions and another under a different set of conditions. By including both of them, a product is obtained which is well adapted for both sets of conditions. It would naturally be expected that the best proportions would vary according to the type of engine and service in which the fuel is to be used, and Bartholomew's application (88) states that this is the case. The supposed invention, therefore, consists merely in combining compounds found to be effective under different conditions, to produce one which will be effective under all conditions. This involves no more invention than combining a compound which will kill mosquitoes with one which will kill flies to produce a preparation which will kill both, or making a mixture of salt and pepper to produce a combined flavoring material. The proportioning of the ingredients will be determined by experiment and will depend on the results desired. This is not a matter involving invention (*Allen* v. *Coe,* 135 F. (2d) 11, 77 U. S. App. D. C. 324).

Bartholomew's contribution was clearly one of routine experimentation rather than one of invention. Knowing that various combinations of lead alkyls were effective as antiknock compounds, and that the particular ones here claimed were effective individually, he merely found by experiment that, used together, they produced, as would have been expected, an over-all result superior to that of any one of them alone. If this is patentable, then the first experimenter to try any new grouping of old compounds which produces a somewhat improved product, is entitled to a patent. This clearly is not what is contemplated by the patent statutes.

It follows that the appealed claims would involve no invention even if the claimed compounds produced some entirely unexpected superiority. A procedure which it is obvious to try cannot be made patentable by the fact that the result is better than would have been expected (*In re Gaureke,* 86 F. (2d) 330, 24 C. C. P. A. 725; *Naamlooze et al* v. *Coe,* 132 F. (2d) 573, 76 U. S. App. D. C. 313; *Minnesota Mining and Mfg. Co.* v. *Coe,* 99 F. (2d) 986, 69 App. D. C. 217). As was said in the last-cited decision:

> But a showing of great industry in experimental research is not in itself sufficient to constitute invention, when the product thereof differs from those of the prior art only in degree and the result, no matter how useful it may be, is merely one step forward in a gradual process of experimentation.

In the present case, moreover, there is clearly no evidence which would justify a holding that the claimed compounds are unexpectedly superior to those of the prior art. No evidence of comparative tests has been submitted to show superiority over the products of the references, or even over the conventional lead tetraethyl. The evidence contains nothing more than general conclusions drawn by the inventor himself to the effect that he has produced a superior product.

Further, even if it could be held that the specific preferred compound disclosed in the application (101) is particularly effective, this would not warrant the allowance of the appealed claims, which are not limited to this compound. On the basis of his alleged discovery that one particular proportioning of certain compounds is useful, Bartholomew seeks to prevent others from using these compounds in any proportions whatever. There is clearly no evidence that every conceivable proportioning of these compounds would produce a useful product and it is thus evident that, even if Bartholomew has made an invention, the claims do not point it out but merely seek to exclude others from the entire field in which it lies.

It is elementary that novelty alone is not sufficient to impart patentability to a claim (*In re Lincoln et al.*, 126 F. (2d) 477, 29 C. C. P. A. 942). Accordingly, the fact, if such it be, that the particular lead alkyls claimed by Bartholomew had not previously been used together, does not warrant the allowance of the appealed claims. It must appear that these substances, when used as set forth in the claims, will produce an unexpectedly improved result. This has not been done.

NO NEW METHOD INVOLVED

The eleventh finding of fact of the Trial Court was that claims 1 to 4 do not define new methods of engine operation. This is clearly correct. The engine operates in exactly the same manner, regardless of the particular antiknock agent used. The inventor (65 et seq.) testified that he does not eliminate knocking, but merely reduces it. Obviously reducing the knocking does not produce a new method of engine operation. If it did, it would be equally logical to say that the use of an automobile using the compound produced a new method of transportation.

The present situation is similar to that in *Ethyl Gasoline Corp. et al.* v. *U. S.,* 309 U. S. 436, in that on the basis of an alleged invention of an antiknock compound, claims have been presented to the compound per se (claims 15 and 16) to a fuel containing the compound (claims 5 to 12) and to a method of engine operation consisting in using this fuel in the conventional way (claims 1 to 4). The purpose of the last two groups of claims is obviously to extend the appellant's control beyond the compound itself, to include the fuel in which it is used, and even the use of such fuel by the ultimate consumer. This practice was condemned by the Supreme Court of the United States in the case last cited, and claims which are evidently designed to implement such a practice should not be allowed.

EVIDENCE SUPPORTS HOLDINGS

It is suggested in appellant's brief that there is "no evidence to support the judgment of the District Court that the subject matter was unpatentable" over the references. Obviously, no evidence other than the patents themselves is necessary to support this holding. If the patents disclose the alleged invention claimed, or if they so closely approach it as to negative patentability, no further evidence is necessary. There is, moreover, Bartholomew's admission (46) that the operation of the particular lead alkyls included in his claims, under conditions of 100° vaporization, "was fairly well known." There is clearly sufficient evidence to support each of the Trial Court's findings.

ALLOWANCE OF OTHER CLAIMS IMMATERIAL

In appellant's brief (page 13) reference is made to two unidentified "recently issued" patents, and it is stated that counsel for the appellee "frankly told this Court in oral argument" that these patents relate to the same general subject matter as the appealed claims, and that they "contain broader claims than those here at bar." Apparently the patents referred to are Campbell 2,304,883 and Smyers et al. 2,310,376. These patents are assigned to General Motors Corporation and Standard Oil Development, which own the Ethyl Gasoline Corporation, the assignee of the Bartholomew application here involved. No attempt has been made by the appellant to place these patents in evidence and accordingly it is not thought that they can properly be considered for any purpose.

Moreover, the appealed claims must be considered on their own merits, and the allowance of allegedly similar or broader claims elsewhere is not controlling (*Dyer* v. *Coe,* 125 F. (2d) 192, 75 U. S. App. D. C. 125; *Sharp* v. *Coe,* 75 U. S. App. D. C. 118; 125 F. (2d) 185; *In re Greider et al.,* 29 C. C. P. A. 1079, 129 F. (2d) 568).

CONCLUSION

It is submitted that all the appealed claims are unpatentable and that the action of the District Court in dismissing the complaint was proper and should be affirmed.

Respectfully submitted.

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REPLY BRIEF FOR APPELLANT

IN THE

United States Court of Appeals

DISTRICT OF COLUMBIA.

No. 8760

ETHYL GASOLINE CORPORATION, *Appellant*,

v.

CONWAY P. COE, and/or his successor in office,
Commissioner of Patents, *Appellee*.

Appeal from the District Court of the United States for the District of Columbia.

UNITED STATES
COURT OF APPEALS FOR THE
DISTRICT OF COLUMBIA

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REPLY BRIEF FOR APPELLANT.

STATEMENT.

The substance of appellee's Brief is that the judgment of the District Court should be affirmed because:

1. The Patent Office has not held the appeal claims patentable over the prior art.

2. The claims are extremely broad.

3. The use of the materials claimed is obvious in view of the prior art.

4. Claims 1-4 do not define a new method.

5. The evidence supports the holdings of the Trial Court.

6. The allowance of claims in other patents is not relevant here.

We shall reply to these points in the order in which they are presented. But, before doing so, we submit that the only issue now before the Court is whether applicant's claims define invention over the four cited patents. We have argued the question of patentability in our main Brief and shall not here reiterate that argument. We have also described and defined the invention of Bartholomew beginning at Page 6 of our main brief. This Court in its first opinion in this case, 139 Fed. (2d) 372 (December 6, 1943), correctly and succinctly defined the invention in the first paragraph of its opinion. That opinion was written after the case had been fully briefed and argued. The invention was carefully described to the Court in oral argument and the opinion discloses that the Court understood the invention and grasped the distinction between the claims involved and the state of the art prior to the existence of the claims in issue.

The essence of appellee's argument now is that appellant is not entitled to a patent because the materials claimed were obvious in view of the prior art. Of course, that prior art, consisting of the four cited patents, was before this Court on the first appeal. The Court did not then hold that such prior art constituted a bar to the allowance of a patent to the appellant.

REPLY TO ARGUMENT OF APPELLEE.

1. Appellee States That the Patent Office Has Not Held the Appealed Claims Patentable Over the Prior Art.

The history of the proceedings in the Patent Office shows that the Patent Office Examiner and the Board of Appeals of the Patent Office held, in their final opinion, that the claims in issue were not patentable over a claim which had been suggested by the Examiner for interference purposes.

That was the sole ground for rejecting the applicant's claims. In both the finding of the Examiner and the opinion of the Board of Appeals the four cited patents constituted a part of the record. It is the well established practice under Patent Office Rule No. 65 that the reasons for the rejection of a claim or claims must be fully and precisely set forth by the Examiner. The reason for the rule is apparent and it is a salutary one. Without such a rule no applicant would ever know the ground upon which his claims are rejected, or when the grounds for rejection might be finally exhausted and the proceedings in the Patent Office come to an end. The rules of procedure in the Patent Office, when not in conflict with any provisions of law, have the full force and effect of statutes. *Mell* v. *Midgley,* 31 App. D. C. 534; *In re Mraz,* 36 App. D. C. 435; *Westinghouse Traction Brake Co.* v. *Christensen,* 243 Fed. 901; *In re Horton,* C. C. P. A., 58 Fed. (2d) 682; *United States ex rel. Steinmetz* v. *Allen,* 192 U. S. 543, 556. The Patent Office having held that Bartholomew was not entitled to a patent because, *and only because,* of his failure to make the suggested claim, thereby ruled in effect that the four cited patents did not constitute prior art against his claims. That this is so is emphasized by the fact that the four items of prior art were briefed by the Examiner before the Board (App. p. 124), and that the Board, while mentioning them (p. 132) held that the disclaimed claim, alone, was the ground of denial of the patent, saying (p. 132) that "it was not intended to raise any new ground of rejection in our decision".

2. The Appellee States That the Claims are Extremely Broad.

The claims are specific to a *primary anti-knock* agent comprising the three compounds named as *primary,* which simply means that one skilled in the art must look to these three compounds to give the anti-knock agent its real effectiveness. The form of the claims was at first objectionable to the Examiner as being too broad or indefinite (App. 117). They were amended by the applicant (App. 118). Thereafter the Examiner held:

> "The claims appear to be in proper form as amended and this ground of rejection is withdrawn." (App. 120)

It is obvious that the experts in the Patent Office including the Board of Appeals, have finally held that the claims are definite and properly define the invention. There is, therefore, no merit in appellee's contention to the contrary.

3. Appellee States That the Use of the Materials Claimed is Obvious in View of the Prior Art.

We have defined Bartholomew's invention beginning at Page 6 of our main brief, and show wherein his claims are wholly different from the disclosures of the cited patents. Among other things, the appellee states in his brief, Page 9:

> "It is obvious that both Sullivan et al and Voorhees contemplate the selection of such of the compounds as are considered desirable."

The fact is that Sullivan et al. and Voorhees make no such statement. Sullivan states that his invention relates to processes of preparing hydro-carbon compounds of lead and it comprises a process wherein a hydro-carbon, for example hexone, is disassociated by the action of an electric spark to liberate free alkyl radicals such as methyl, ethyl, propyl or higher alkyl radicals, and these reaction processes are caused to combine with lead whereby lead alkyl compounds are formed. There is no discussion whatever in the patent as to which of the alkyls, or combination of alkyls, is desirable for any purpose other than anti-knock properties generally. He gives no directions as to the forming of mixtures or the value of the different mixtures (App. 163).

Voorhees discloses a method of treating sodium chloride and lead to make tetra-ethyl lead or the like, and to apply it to gasoline. He says:

> "Although I am not prepared to state the exact nature of the lead-hydrocarbon compounds in my gasoline I believe that they are more complex in chemical structure than the simple alkyl compounds and the hydro-carbons contained in general at least four carbon atoms." (App. 170, line 97.)

Thus the cited patents specifically referred to by the appellee to substantiate his contention entirely negative it.

In our main brief, at pages 6 *et seq.*, we pointed out Bartholomew's problem and the fact that all known thought processes, tests, and routine experimentation had been exhausted and the problem remained unsolved. Bartholomew conceived what he thought would be a solution of the problem. He constructed new test apparatus and charts in order to confirm his conception and prove his invention (App. 29, 30, 31, 44-49, 66). The reading of Bartholomew's testimony, his application, and the claims, completely explodes the appellee's assumption that the use of the materials claimed is obvious and that Bartholomew's work was purely routineering, and that his result was obtained by experimentation. The fact is that no amount of experimentation or routineering would ever have solved the problem. New thought processes, new test apparatus, and new charts were essential. All of these were essential to Bartholomew's original concept and invention. His was: ". . . a new display of ingenuity beyond the compass of the routineer". *Kirsch Manufacturing Co.* v. *Gould Mersereau Company, Inc.*, 6 Fed. (2d) 793, 794. His claims come within the principle recognized by this Court in *Dean Murphy et al.* v. *Coe*, 99 Fed. (2d) 995.

> "In *Becket* v. *Coe*, 60 App. D. C. 53, 98 Fed. (2d) 332, combinations of some five metals, in given proportions, to form a stain-resisting and deep-drawing alloy, were held patentable notwithstanding the fact

> that the metals and the proportions came within ranges previously disclosed by others."

Appellee's Brief is obviously directed to the fallacy that all Bartholomew did was to experiment or routineer. Clearly that is not correct.

4. Appellee States That Claims 1-4 Do Not Define a New Method.

His argument on this point is set forth at Page 14 of his Brief. We have answered this argument at Page 15 of our main brief.

5. Appellee States That the Evidence Supports the Holdings of the Trial Court.

His argument in support of this contention is at Page 15 of his Brief. Again his principal reliance is on the four patents. We have discussed this question in our main brief beginning at Page 12. We reiterate that there are no facts in the record, including the patents, which in any way support the holding of the Trial Court to the effect that these claims are unpatentable over the cited patents.

6. Appellee Further States That the Allowance of Claims in Other Patents is Not Relevant Here.

Our position is that the appellee told the Court on the former hearing that the Patent Office has issued two patents on the same general subject matter as contained in the claims here in issue. Those patents were issued to parties who made the suggested claim and entered into the interference proceeding proposed by the Examiner in the Patent Office. They were issued during the pendency of this litigation. This Court has held that Bartholomew did not disclaim his invention by refusing to make the suggested claim. We concede that the allowance of claims in other patents is not determinative of patentability here, but feel confident that it does negative the appellee's argument

(Brief, p. 5) that "if the subject-matter of the proposed claim be treated as prior art, the appealed claims are unpatentable thereover."

A MISTAKE IN APPELLEE'S BRIEF.

The statement is made at page 14 of our adversary's brief that the present situation is similar to that in *Ethyl Gasoline Corporation* v. *United States,* 309 U. S. 436, in that the claims cover the compound *per se,* a fuel containing the compound, and a method of engine-operation consisting of using this fuel in the conventional way. To that is appended the statement that "This practice was condemned by the Supreme Court of the United States in the case last cited, . . ." Our adversary does not point out the basis for this statement, and upon reading the opinion we do not find any basis for it.

CONCLUSION.

The judgment of the Court below should again be reversed, and the appellee should be authorized to issue a patent to the appellant.

Respectfully submitted,

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